PCDD/F IN SOILS AND FREE-RANGE CHICKEN EGGS IN VIETNAM: LONG-TERM IMPACT OF AGENT ORANGE SPRAYING

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Introduction

About 15 % of the area of South Vietnam's forests have been sprayed once, and additional 4 % have been sprayed multiply with herbicides[1]. In addition to spraying two major spills of Agent Orange and Agent White had occurred in December 1969 and 1st March 1970 in Bien Hoa Airbase[2]. Minh et al.[3] showed that Agent Orange has remained the primary source of dioxins in Bien Hoa airbase. A food consumption frequency survey of 400 households in Bien Hoa in 2007 showed that 10 % of households raised poultry, cattle or grew crops and vegetables[4]. Hens foraging on soil contaminated with PCDD/Fs, even at low levels (ppt), can accumulate these compounds to an unacceptable concentration. From 5 to 30 % (mean 28 %) of the intake of dioxins from soil is excreted into the eggs[5]. Feeding hens can ingest 2-30 g of soil with fodder depending on soil coverage and the available surface area per chicken in the outdoor run[6]. The uptake of contaminants in soil and its carry-over to eggs depend on several factors, such as the concentration of the contaminant in the soil, bioavailability, metabolic stability, flock size, foraging behavior, time spent outdoors, and accessibility of feed[7-9]. The age of the hens also influences the level of dioxins found in eggs[10].

The aim of this study was to assess the long-term impact of Agent Orange spraying on PCDD/F levels and congener profiles found in free-range chicken eggs and soils.

Materials and methods

Samples were collected from private households from different Vietnam provinces. A large territory from Lao Cai province in the north to Dong Nai province in the south was covered, including a known dioxin hotspot - the Bien Hoa airport, where Agent orange storage were located during the Vietnam war. A total of 46 chicken egg samples and 36 soil samples were collected and analyzed between 2010 and 2014 years. After sampling all eggs were hard boiled and frozen. Prior to extraction samples were spiked with a mixture of ^{13}C-labelled standards. After the extraction samples were subsequently cleaned up on activated carbon column, multi-layer column and aluminum oxide column. Extracts were then analyzed for PCDD/F using HRGC-HRMS method. Lipid content was determined gravimetrically. Calculation of total TEQ was based on WHO-TEF_{2005}[11]. For values below the detection limit the respective detection limits were used.

Results and discussion

Total dioxin concentration in eggs ranged between 0.4 and 361 pg WHO-TEQ2005 g-1lipid (fig.1). The highest concentration was observed in household near Bien Hoa airport. All samples from the northern part of Vietnam were below the current EC limit of 2.5 pg WHO-TEQ2005 g-1lipid. Whereas PCDD/F concentrations in the majority of samples in the southern part exceeded this value. Total TEQ in soils ranged between 0.1 to 1272 pg TEQ/g (fig.2)

All sampling sites were divided into 3 groups using the map of spraying missions in Vietnam (https://www.chicagotribune.com/chi-091204-agentorange-map-htmlstory.html):

- Southern sprayed with Agent Orange
- Southern not sprayed
- Northern

Total TEQs in chicken eggs statistically significantly ($p<0.05$) differed between all three groups, being higher for sprayed territories and lower for northern. Mean concentrations in eggs were 1.4, 3.3 и 6.7 pg WHO-TEQ g^{-1} lipids respectively. Mean concentrations in soils were 0.4, 1.4 и 4.1 pg WHO-TEQ g^{-1} (fig.3)

It was found that elevated 2,3,7,8-TCDD contribution to total TEQ is characteristic only for southern territories sprayed with AO (fig.4). It amounted to 70 % in eggs and to 80 % in soils (mean values were 50% and 59 % respectively), whereas in northern territories and southern territories that were not subjected to Agent Orange spraying mean TCDD contribution was about 12 % in soils and 14% in eggs.

Although according to Hoang et. al[12] samples collected from the sprayed areas do not really show higher levels than in other locations, our results show not only higher levels in sprayed areas but also a pattern of TCDD domination characteristic for dioxin exposure from Agent Orange.

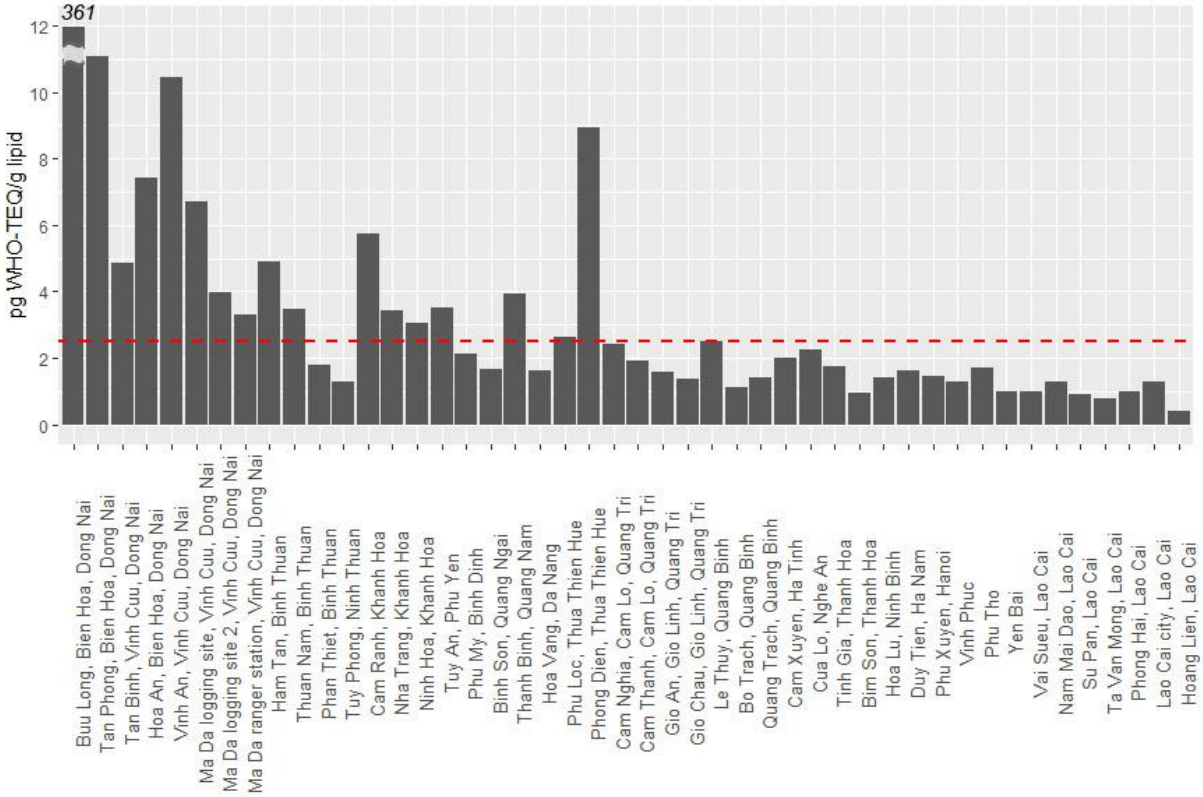

Fig.1. Total TEQ values in chicken eggs from Vietnam arranged from south to north. The red line represents the EC limit for eggs and egg products (2.5 pg WHO-TEQ2005 g-1lipid).

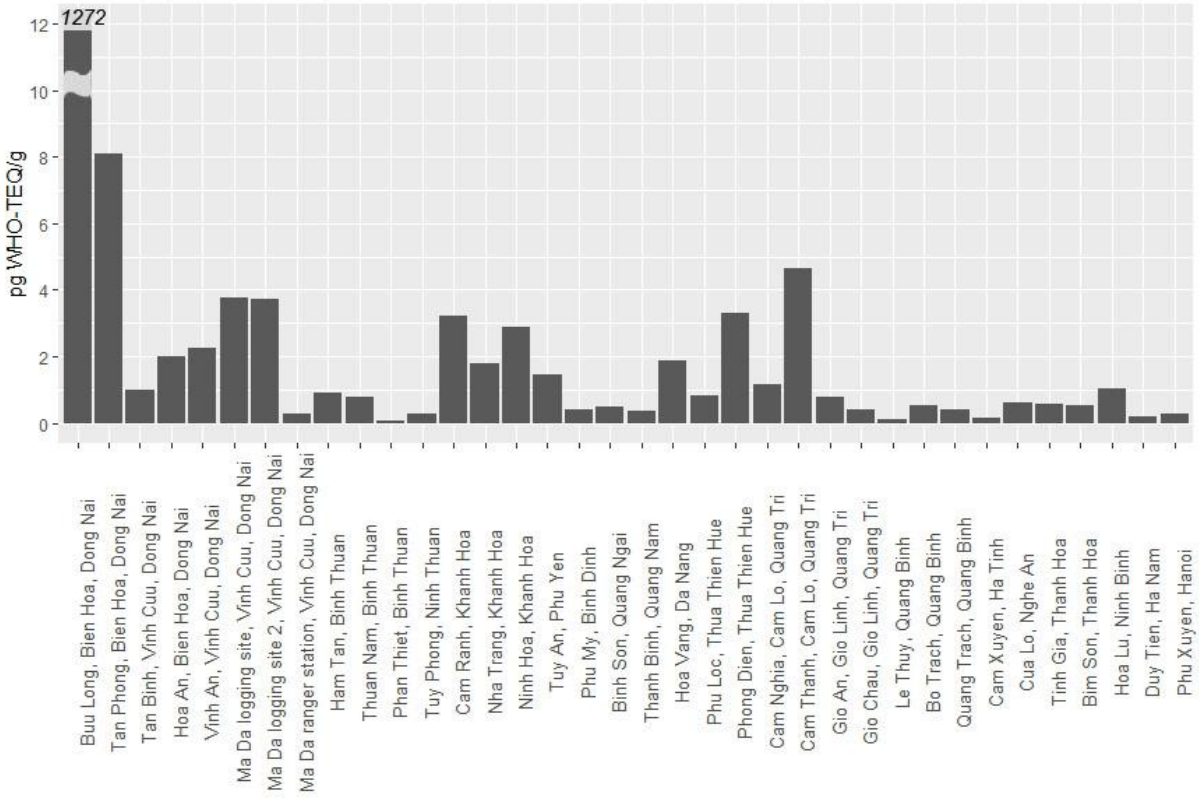

Fig.2. Total TEQ values in soils from Vietnam arranged from south to north.

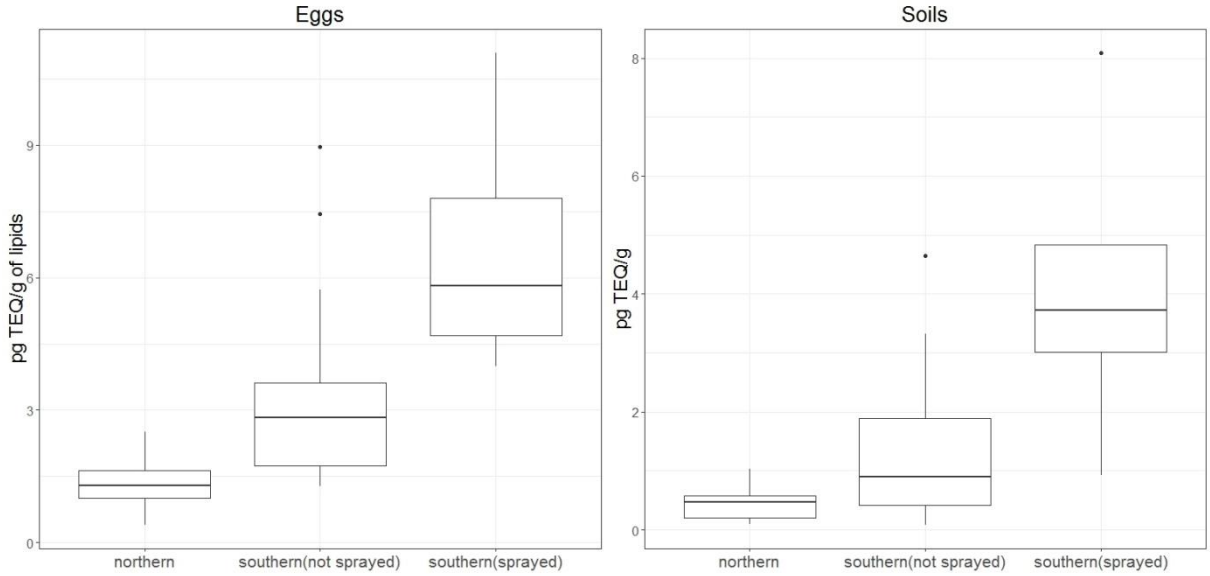

Fig.3. Total TEQ in chicken eggs and soils from northern, southern not sprayed with Agent Orange and southern sprayed sites.

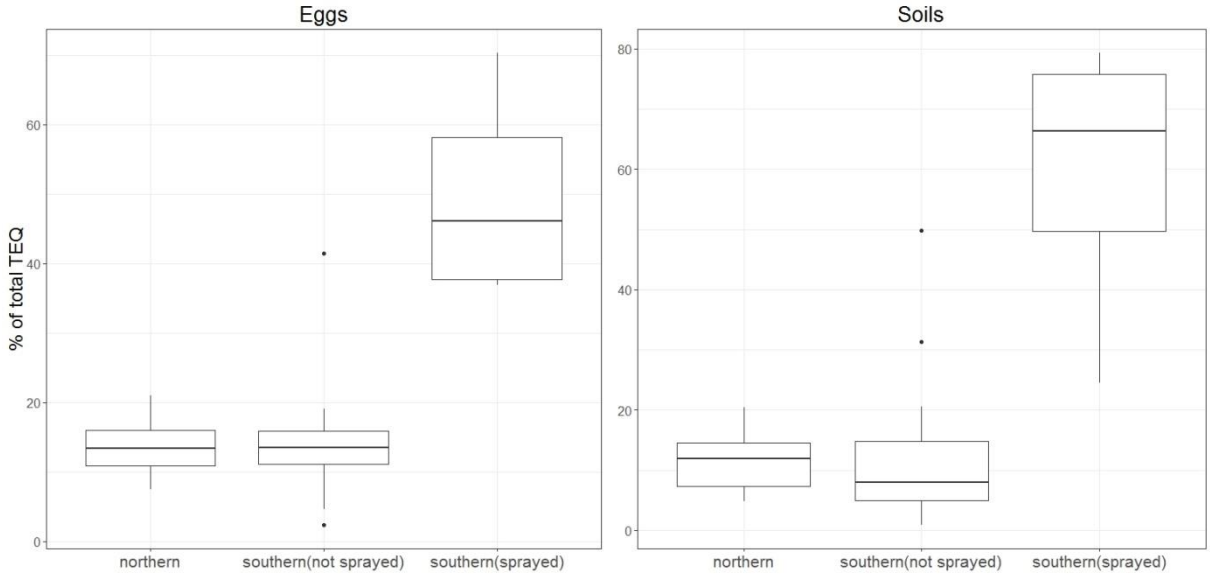

Fig.4. 2,3,7,8-TCDD contribution to total TEQ in chicken eggs and soils from northern, southern not sprayed with Agent Orange and southern sprayed sites.

The mean proportions of the total PCDD/F concentration for all toxic congeners were calculated to assess typical profile for each group (fig.5). In eggs the dominant contributors in all groups except the hot spot were OCDD and HpCDD. The mean proportions of some furans are somewhat higher in northern territories (2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF и OCDF). However, only in case of TCDF the difference is statistically significant ($p<0.05$). Proportion of HpCDD is in contrast higher in southern territories
Similar patterns were not observed in soils. OCDD was the main contributor in all groups.

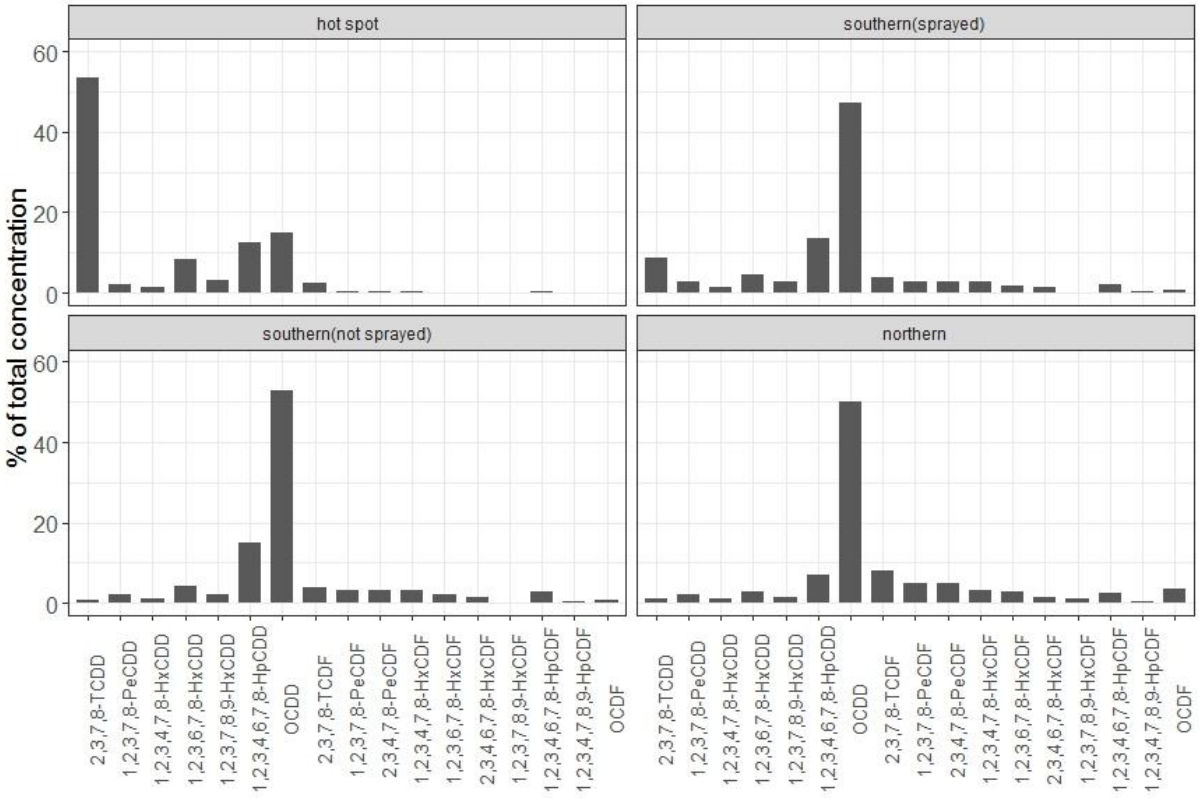

Fig.5. Congener profiles in chicken eggs on total concentration basis

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