POTENTIAL HUMAN HEALTH RISKS CAUSED BY CONTAMINATED FOOD CONSUMPTION EVALUATED AT SITE AFFECTED WITH HERBICIDES SPRAYING; PHONG MY COMMUNE, CENTRAL VIET NAM

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1. Introduction

This study was focused on possible risks caused by PCDD/PCDF contamination of environment and human foodchain to local inhabitants in central Viet Nam. During the war conflict in 1962 through 1971, the US Army forces used about 72 million litres of herbicides (usually mixture of 2,4-D (2,4-dichloro-phenoxy acetic acid) and 2,4,5-T (2,4,5-trichloro-phenoxy acetic acid) and sprayed them over large areas in central and south Viet Nam.

This study was settled to Phong My Commune, Thua Thien-Hue province in central Viet Nam. This site was contaminated by herbicides (predominantly Agent Orange) spraying. At most of the area the herbicides were applied several times.

Over two years (2006 – 2007) 118 samples were taken at site from all main environment and human foodchain matrices. These samples were analysed for PCDD/PCDF content. In the first stage of investigation, other chemicals such as PCB and chlorinated pesticides were analysed as well. However, presence of these contaminants was not identified. Within the scope of the works, an inquiry contained 27 questions focused above all on dietary habits and lifestyle was also organized for the inhabitants of Phong My Commune. In this paper, we report our findings about main human health risks caused through exposure scenario “consumption of contaminated food”.

2. Methods

Studied matrices

Large geochemical investigation oriented on contamination of the environment with PCDD/PCDF, PCB, organochlorinated pesticides and heavy metals was carried out in Phong My commune. In total 118 samples were taken including 39 soil samples, 13 sediment samples, 5 water samples, 57 samples of animal tissues and 4 samples of fruits.

Sampling of animal tissues was focused mainly on fish and poultry, however, samples of pork, beef and wild animals (snake and frog) were collected too. Number of fish species was analysed: Chana Maculata, Cyprinus Carpio, Caraassius, Clarias Macrocephalus etc. Within the poultry sampling meat of chickens, hens, cocks and ducks were analysed.

Soil and sediment samples were collected representatively from the whole area of the commune, predominantly from the area of animal breeding and fish farming. Samples of groundwater and surface water were collected from the existing drinking water sources.

Methods of PCDD/PCDF analysis

The method of composite samples was used for collecting all soil samples. Samples of soil, water, animal tissues, fruits, vegetables and human blood were taken as a single sample. The samples were analyzed in the specialized ALS dioxin laboratory in the Czech Republic. An ALS standard analytical procedure according to the United States Standard EPA 1613 and US EPA 668 for determination of field chlorinated dibenzo-(p)-dioxins and dibenzofurans, was used. The soil and sediment samples were extracted with organic solvents (toluene or acetone/toluene mixture depending on the dry matter content) using ASE-300 apparatus. The obtained raw extract was pre-cleaned by multiple column chromatographs in order to eliminate interference with co-extracts. After concentration and spiking with injection standard, a fraction of the final extract was analyzed by HRGC-HRMS.

The separation of each 2,3,7,8 PCDD/PCDF was assured by a column with polar stationery phases and the detection was assured by a mass spectrometer working in the MID operating conditions with high resolution (R≈10 000). Quantitative analysis was performed using selected ion current profile (SICP) areas. For determination of the analytes, the HRGC-HRMS system was calibrated and the concentration of each compound was determined by an isotope dilution technique using certified 13C12 2,3,7,8-PCDD/PCDF standards with response factors. Relative response factors were determined using five-point calibration (CS1-CS5) for each group of the contaminants.

Human health risk assessment approach

The calculations of exposure doses, carcinogenic and non-carcinogenic health risks were carried out according to the standard methodics for calculating exposure and health risks using exposure parameters recommended by the methodologies of US EPA[1] and the Ministry of Environment of the Czech Republic[2]

According to several elaborated studies, including the US-EPA study, the recommended tolerable daily intake (TDI) of substances with dioxin effect (PCDD, PCDF, and coplanar PCB) ranges currently within the interval 1 – 10 pg $I\text{-}TEQ_{Total}$ per kg of body weight[3], where $I\text{-}TEQ_{Total}$ is internationally used toxic equivalent of the PCDD/PCDF/PCB mixture relative to TCDD. The Agency for Toxic Substances and Disease Registry[4] determined minimum risk level of exposure to dioxins at 1 pg I-TEQ per kg of body weight and day. The Health Council of the Netherlands (1996) recommended the tolerable daily intake for the group PCDD/PCDF/PCB at 1 pg $I\text{-}TEQ_{Total}$/kg of body weight. This value was based on immunological effects and nervous system defects resulting from chronic dietary exposure of primates (macaques and marmosets). This was the very first evaluation in which compounds with dioxin effects, such as PCB, were also included into TDI[3]. According to the Scientific Committee on Food[5] the value of tolerable weekly intake TWI 14 pg WHO-TEQ/kg of body weight was recommended. This value falls within the range of TDI 1 – 4 pg WHO-TEQ/kg of body weight determined based on consultations of WHO (World Health Organization) and IPCS (International Programme on Chemical Safety) in 1998 for substances with dioxin effect. One of main conclusions of this consultation was also specification of the objective to reduce exposure of man to dioxins below 1 pg WHO-TEQ/kg of body weight and day[3].

In case the threshold effect of dioxins is not considered and genotoxic effects leading to development of tumour diseases are taken into account, the tolerable level of exposure to these substances becomes even much lower. An example can be TCDD risk evaluation[6] that, based on genotoxic effect of this substance on animals, determined the tolerable daily intake 5 – 50 fg TCDD/kg of body weight. This value also corresponds to conclusions of US EPA (Environmental Protection Agency), that determined, based on human epidemiological studies in 2000, the upper limit of the carcinogenic risk slope at 1×10^{-3} pg TCDD/kg of body weight and day in lifetime exposure to 2,3,7,8-TCDD. The quantitative estimate of the risk of tumour disease development (carcinogen risk slope CSF) for peroral as well as inhalation intake of 2,3,7,8-TCDD referred to in the Risk Based Concentration Tables[7] corresponds to 1.5×10^{5} $(mg/kg/d)^{-1}$.

The toxicological database RAIS[8] gives the values of carcinogen risk slopes $1.16x10^{5}$ $(mg/kg/d)^{-1}$ and 3×10^{5} $(mg/kg/d)^{-1}$ for the inhalation and dermal exposure pathways, respectively. For the contaminant 2,3,7,8 TCDF, the stated values of slopes for peroral, inhalation and dermal ways of exposure are by an order of magnitude lower, CSForal = 1.5×10^{4} $(mg/kg/d)^{-1}$, CSFinhalation = 1.16×10^{4} $(mg/kg/d)^{-1}$, and CSFdermal = 3×10^{4} $(mg/kg/d)^{-1}$ (RAIS 2007).

3. Results and Discussion

PCDD/PCDF contamination

The contents of the most toxic (2,3,7,8 substituted) congeners of PCDD/PCDF in the collected soil and sediment samples are relatively low and ranged within the interval of 0.05 – 5.1 pg I-TEQ/g dw. The highest PCDD/PCDF contents were found in case of soil from non-cultivated areas – hillsides where the dioxin content was 5.1 pg I-TEQ/g dw. Average content of PCDD/PCDF in the soil samples was 2.13 pg I-TEQ/g dw (calculated as maximum possible value of I-TEQ, so-called „Upperbound“). The most often found congeners in soil are

2,3,7,8–TCDD and OCDD, wherein the proportion of the most toxic congener 2,3,7,8-TCDD in total I-TEQ was 14 % on average (participation of 2,3,7,8-TCDD on total I-TEQ increases with the distance from the settlements). If the analyses where 2,3,7,8–TCDD was detected are only taken into consideration, its proportion in the total I-TEQ is 55 % on average. This proportion is however lower than that stated in a comparable study realized in the neighbouring district A Luoi by Hatfield Consultants[9] where the proportion of 2,3,7,8-TCDD in soil samples ranged within the interval of 86 – 94 % of total I-TEQ. Generally, soil samples (except those from house close neighbourhood) are characterized by a very low or zero content of furans (PCDF), which indicates the main source of contamination from herbicide sprays, primarily Agent Orange.

Contents of toxic PCDD/PCDF congeners in the fish samples ranged between 0.09 and 4.8 pg/g of sample WHO-TEQ; average content of PCDD/PCDF was 0.8 pg/g of sample WHO-TEQ. Except one fish catched in the dam, PCDD/PCDF concentrations in the all other fish samples did not exceed the EU limit content of dioxines in fish meat and fish products, which is 4 pg/g fresh weight WHO-TEQ. The highest concentrations were found in the fish species Chana Maculata and Clarias Macrocephalus netted in the dam. In the dioxin analyses in fish, recalculation to the lipid basis proved to be principal. Average content of PCDD/PCDF in the fish adipose tissue was 37.5 pg/g of fat WHO-TEQ.

In the poultry samples, the ascertained values of PCDD/PCDF ranged within the interval 0.09 – 4.8 pg/g of sample WHO-TEQ; after recalculation to the fat tissue, the interval was 1.1 – 14 pg/g of fat WHO-TEQ. In most cases, the found contents were above the limit of the European Directive EC No. 199/2006 (2 pg/g of fat). Average value of WHO-TEQ PCDD/PCDF was 0.8 pg/g of sample and 5.3 pg/g of fat. From the congener distribution, a dominant proportion of 2,3,7,8-TCDD in total WHO-TEQ is apparent.

PCDD/PCDF contents in pork and beef samples were relatively low. In case of pork meat, average PCDD/PCDF content was 0.36 pg/g of sample WHO-TEQ, and 0.55 pg/g of fat WHO-TEQ, respectively. In case of beef meat, average content was 0.51 pg/g of sample WHO-TEQ and 1.3 pg/g of fat WHO-TEQ, respectively.

Human health risk evaluation

With respect to the determined concentrations of PCDD/PCDF in food, above all in fish and poultry, it was considered that eating of this food is a realistic exposure scenario. Most hazardous foodstuffs are mainly fish, poultry, wild animals (snakes, frogs etc.). An absolute majority of foodstuffs, being consumed in the given area, is being grown directly in place. Substantial part of inhabitants is self-sufficient in food production. It resulted from the inquiry that the diet habits in the examined groups of inhabitants do not differ much, and this exposure scenario can be considered as valid for all local inhabitants.

Overall maximum concentrations in the average food sample of the local inhabitants show the average daily intake 13.2 pg/kg/day WHO-TEQ in adults (18 – 70 years), which is a value approximately 5-times to 7-times higher in comparison with estimated exposure doses from food in Europe and North America. The highest value of average daily dietary intake was found in the age category 10 to 18 years at the level of 17.7 pg/kg/day WHO-TEQ. In case average concentrations found in food samples were used for calculating exposure the average daily dietary intake PCDD/PCDF for monitored age categories ranged between 4.6 and 6.1 pg/kg/day WHO-TEQ. In many European countries, the average daily dose of PCDD/PCDF after recalculation to the total equivalent toxicity TEQ ranges within the interval 1.5 – 2 pg/kg of body weight. In the USA and Canada, the estimated daily intake PCDD/PCDF ranges within the interval of 1 - 3 pg/kg of body weight. In case the average daily dose includes also equivalent toxicity of coplanar (so called dioxin-like) PCB the daily intake in Europe and USA results in 2 – 6 pg/kg of body weight WHO-TEQ[10]. In case of the monitored population in Phong My Commune, the following proportion was found of respective food components on the total intake of PCDD/PCDF through dietary exposure: (73.8 % fish meat, 19.0 % rice, 4.3 % poultry meat, 1.6 % other meat, 0.8 % vegetables, 0.5 % fruits). This profile was similar in all the monitored age categories from 1 to 70 years.

According to the study in Italy[11] it is, for example, it is possible to expect two-times to three-times higher differences in the dietary intake between various age categories (0-6 years, 7-12 years, and 13-94 years) (with a maximum found in small children) depending on dietary habits of the respective groups. The results also indicate that the highest contribution to exposure to PCDD/PCDF and, possibly, to coplanar PCB is connected with consumption of fish and dairy products. According to the study carried out in the Netherlands[12], a significant contribution to PCDD/PCDF/PCB exposure comes also from consumption of meat products, vegetables, and eggs making 23 %, 13 %, and 4 % proportion, respectively, in the total exposure to these pollutants from food.

Results of the model calculations proved that assumptions connected with the exposure scenario “consumption of contaminated food” had been justified and non-carcinogenic as well as carcinogenic risks for all studied age categories of inhabitants connected with the dietary exposure were confirmed. The found out values of the hazard index HI for the non-carcinogenic risk of the monitored age categories range between 13.3 and 17.7 for maximum PCDD/PCDF concentrations in foodstuffs; whereas the ILCR values (lifetime increase in probability of tumour disease development) for the investigated population groups ranged between $2.8.10^{-5}$ and $1.5.10^{-3}$; this corresponds to the probability of tumour disease development approximately within the interval of 28 individuals from the group of one million inhabitants to 15 individuals from 10,000. The cumulative lifetime risk of tumour disease development in the monitored population corresponds to the value $2.1.10^{-3}$ for the maximum PCDD/PCDF concentrations found in foodstuffs.

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