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Environmental fate and dietary exposures of humans to TCDD as a result of the spraying of Agent Orange in upland forests of Vietnam

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HIGHLIGHTS

- We simulate exposure to TCDD resulting from spraying of Agent Orange in Vietnam.
- Modeled body burdens were broadly consistent with available biomonitoring data.
- Potential exposures in 'spray zones' are relatively small compared to 'hot spots'.

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ABSTRACT

The fate and transport of 2,3,7,8-tetrachloro-p-dibenzodioxin (TCDD) released into the environment of South Vietnam (SVN) as a consequence of the aerial application of the herbicidal defoliant Agent Orange (AO) were simulated for a generic upland forest scenario and followed over a 50-year period (1965, 1968 and 1970 onwards). Modeled concentrations of TCDD in the environment were then used as inputs to a human exposure model, which focused on long-term exposures via the food chain. Intake rates and body burdens of TCDD were estimated for adult males over the course of the simulation period and compared to available biomonitoring data. One of the most important factors determining the magnitude of the simulated human exposure to TCDD was the fraction of the chemical deposited directly to soil (where it was assumed to have a degradation half-life of 10 or 15 years) relative to the fraction assumed to remain on/in the forest canopy following the spray application (where it was assumed to have a degradation half-life of ≤48 h). The simulated body burdens under the various scenarios considered were broadly consistent with the biomonitoring data from SVN collected in the mid-1980s to late 1990s. Taken together, the modeling results and empirical data suggest that highly elevated exposures to TCDD (i.e., body burdens in the several 100s of pg/g lipid range and greater) were not common among people inhabiting upland forest locations in SVN sprayed with AO and that peak and average body burdens were broadly similar to those of the general population of the U.S. in the 1970s and early 1980s. The model-based assessment is consistent with the 'hot spot' hypothesis i.e., potential exposures to TCDD linked to activities conducted on or near former bases where AO was stored are greater than potential exposures in areas subjected to aerial spraying.

1. Introduction

The most widespread use of herbicidal defoliants in wartime was in South Vietnam (as it was known at the time) during the Second Indochina War (1961–1975) (Westing, 1984a). The use of these herbicides generated considerable human and environmental exposure concern from the public and this was followed by a large number of scientific studies, panel reviews, and books. The findings

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have been cataloged in publications such as Westing (1984b) and the reports of the National Academy of Sciences on the use of Agent Orange (AO) during this conflict (National Academy of Sciences, 1974, et seq.).

Many of the papers and scientific panels that have addressed the use of defoliants in South Vietnam (SVN) have focused on potential adverse effects on human health with special emphasis on the dioxin contaminants found in some of the herbicides used for the purpose of defoliation (IOM, 2014). This paper presents a critical analysis of long-term exposures to dioxins, specifically 2,3,7,8-tetrachloro-p-dibenzodioxin (TCDD) from the environment, and how these exposures may have resulted in movement through the food chain and ultimately in exposures to humans (i.e., Vietnamese civilians). In a companion paper we considered direct human exposure pathways for U.S. military personnel including contact with spray and post application contact with sprayed foliage (Ross et al., 2014).

Although the mixtures of herbicides used in the operations in SVN contained several dioxins, the dioxin of major interest is TCDD because it is by far the dominant congener present in AO (and other 2,4,5-T formulations) (Rappe et al., 1978) and it is the most toxic to mammals (Safe, 1998). In this analysis, consideration was given to environmental processes that result in dissipation and degradation of TCDD and how these may have affected persistence in the environment and movement into the food chain. Modeling was used and the outputs of the models have been compared to historical and recent measurements of concentrations of TCDD in the environment and humans from areas that were sprayed with herbicides.

As part of the campaign in SVN, spraying of defoliants began in 1962 and ceased in 1971 (Westing, 1984a). During this period, 72 million L (Westing, 1984a) to 74 million L (Young, 2009) of herbicides was sprayed over an area of 1.6 to 1.3 million ha (Westing, 1984a; Young, 2009), with AO (a mixture of 2,4,5-T and 2,4-D) accounting for approximately 60% of the total herbicide spray volume (and >96% of the 2,4,5-T herbicides containing TCDD) (Stellman et al., 2003) followed by Agent White (a mixture of 2,4-D and picloram) and Agent Blue (a mixture of dimethyl arsenate and dimethylarsinic acid). Most of this area (1.125 million ha) was sprayed only once while 0.382, 0.136, 0.048, and 0.019 million ha were documented as being sprayed 2, 3, 4, and ≥5 times, respectively (National Academy of Sciences, 1974; Westing, 1984a). The results of this spraying were that large areas of land were denuded of broadleaf vegetation as well as grasses that are susceptible to damage from the applied herbicides. Of the total volume of AO applied, approximately 90% was used on forests (or 'miscellaneous woody vegetation') with the remainder used to target susceptible crops (Westing, 1984a). Some tree species were only damaged if they experienced multiple spray exposures, with limited damage from single applications. Although the killing of the vegetation itself resulted in major ecological effects on communities of plants (Van Trung, 1984) and animals (Quy, 1984) in the sprayed areas, these ecological effects were not included in our assessment. We focus on AO because only herbicide mixtures containing 2,4,5-T were contaminated with TCDD and AO use was by far the largest in terms of volume applied and area sprayed. The justification for focusing on upland forests is that this type of vegetation was targeted most frequently (e.g., 5 800 000 ha of dense upland forest sprayed vs. 500 000 ha of mangrove forest) (Westing, 1984a).

Problem formulation (PF) is the initial stage of all assessments of risks and brings together the information about the stressor in question (TCDD in this case) and the environment to focus the assessment on the key determinants of exposure and toxicity (IOM, 2014). Thus it starts with the identification of the types of technical analyses needed and finishes with a series of hypotheses to be tested with experimental or modeled data. In this assessment, the sole focus was on human exposure and therefore information has been limited to this objective. The result of the PF was the development of a conceptual model of potential indirect exposure pathways via the food chain. Background information characterizing potential exposure scenarios is provided in the Supplementary information (SI, Section S1).

2. Methods

A diagrammatic illustration of the scenarios and models used to simulate exposures of humans to TCDD from the aerial spraying of herbicides (UC-123 aircraft) over upland forest environments in SVN is presented in Fig. 1. Details on the parameterization and application of the modeling tools are presented in the following sections and SI.

2.1. Fate and transport of TCDD

The fate and transport of TCDD in upland forest environments of SVN were simulated using a modified version of the CoZMo-POP 2 model (Wania et al., 2006). CoZMo-POP 2 is a generic, fugacity-based model (Mackay, 2001) originally developed to describe the behavior of neutral organic chemicals with a focus on quantifying riverine and atmospheric pathways for delivering contaminants to large freshwater lakes or the coastal marine ecosystem. The model was recently applied to simulate the link between environmental emissions, exposure and human body burdens of polychlorinated biphenyls (PCBs) in the Baltic Sea drainage basin (Breivik et al., 2010).

The modeled environment includes vegetation (forest canopy), soils, freshwater bodies and a number of linked marine water units characterizing estuarine, coastal and open-water systems. The fate and

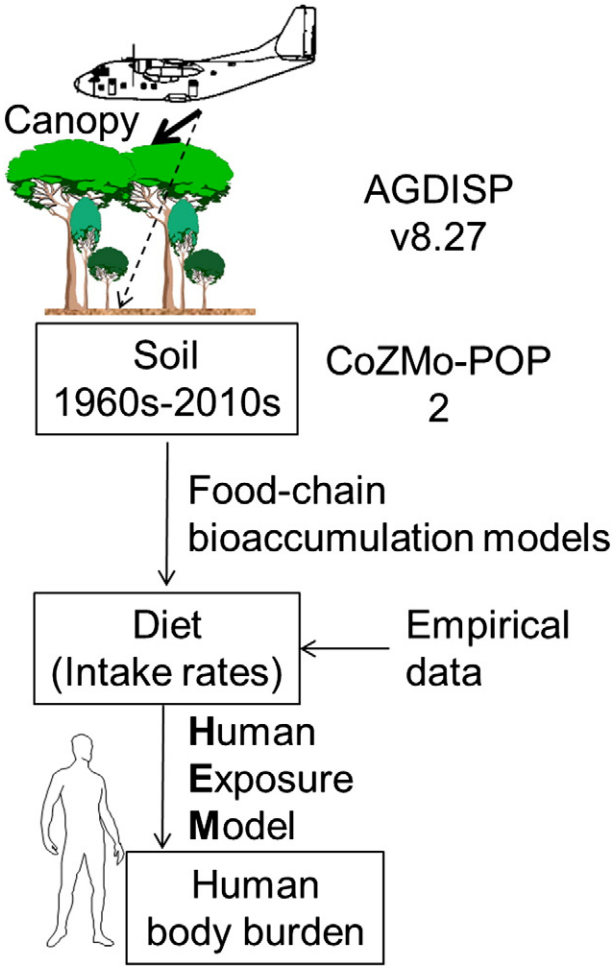

Fig. 1. Diagrammatic illustration of the scenarios and models (AGDISP v8.27, CoZMo-POP 2, HEM) used to estimate exposures of humans in South Vietnam to TCDD from the aerial spraying of herbicides (UC-123 aircraft) over upland forest environments.