



TDS exposure project: Relevance of the Total Diet Study approach for different groups of substances



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ABSTRACT

A method to validate the relevance of the Total Diet Study (TDS) approach for different types of substances is described. As a first step, a list of >2800 chemicals classified into eight main groups of relevance for food safety (natural components, environmental contaminants, substances intentionally added to foods, residues, naturally occurring contaminants, process contaminants, contaminants from packaging and food contact materials, other substances) has been established. The appropriateness of the TDS approach for the different substance groups has then been considered with regard to the three essential principles of a TDS: representativeness of the whole diet, pooling of foods and food analyzed as consumed. Four criteria were considered for that purpose (i) the substance has to be present in a significant part of the diet or predominantly present in specific food groups, (ii) a robust analytical method has to be available to determine it in potential contributors to the dietary exposure of the population, and (iii) the dilution impact of pooling and (iv) the impact of everyday food preparation methods on the concentration of the substance are assessed. For most of the substances the TDS approach appeared to be relevant and any precautions to be taken are outlined.

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Abbreviations: AFM1, aflatoxin M1; BBP, butyl benzyl phthalate; BFR, brominated flame retardant; BPA, bisphenol A; DEHP, di(2-ethylhexyl) phthalate; DiBP, diisobutyl phthalate; DIDP, diisodecyl phthalate; DnBP, di-n-butyl phthalate; DON, deoxynivalenol; FB1, fumonisin B1; FID, flame ionization detection; GC, gas chromatography; GC/ECD, gas chromatography with electron capture detection; GC-MS, gas chromatography–mass spectrometry; GC MS/MS, gas chromatography–tandem mass spectrometry; HBGV, health-based guidance value; HPLC, high-performance liquid chromatography; ICP-OES, inductively coupled plasma optical emission spectroscopy; ICP-MS, inductively coupled plasma mass spectrometry; LC-HRMS, liquid chromatography with high resolution mass spectrometry; LC-MS/MS, liquid chromatography–tandem mass spectrometry; LOD, limit of detection; MOH, mineral oil hydrocarbons; MOAH, mineral oil aromatic hydrocarbons; MOSH, mineral oil saturated hydrocarbons; MRL, maximum residue level; MS, mass spectrometry; OTA, ochratoxin A; PAH, polycyclic aromatic hydrocarbon; PBDE, polybrominated biphenyl ether; PCB, polychlorinated biphenyl; PCDD, polychlorinated dibenzo-p-dioxin; PCDF, polychlorinated dibenzofurans; PFBS, perfluorobutanesulfonic acid; PFDA, perfluorodecanoic acid; PFDoA, perfluorododecanoic acid; PFHpA, perfluoroheptanoic acid; PFHxA, perfluorohexanoic acid; PFHxS, perfluorohexane sulfonate; PFNA, perfluorononanoic acid; PFOS, perfluorooctane sulfonate; PFOA, perfluorooctanoic acid; PFUnA, perfluoroundecanoic acid; POSH, polyolefin oligomeric saturated hydrocarbon; TDS, Total Diet Study; TE, trace element.

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1. Introduction

A Total Diet Study (TDS) generally consists of selecting, collecting and analysing commonly consumed food purchased at retail level on the basis of food consumption data to represent a large portion of the typical diet, processing the food as for consumption, pooling the prepared food items into representative food groups, homogenizing the pooled samples, and analysing them for harmful and/or beneficial chemical substances (EFSA, 2011a). From a public health point of view, a TDS can be a valuable and cost effective complementary approach to food surveillance and monitoring programs to assess the presence of chemical substances in the population diet and to provide reliable data in order to perform risk assessments by estimating dietary exposure. International organisations such as the Food and Agriculture Organization of the United Nations (FAO), the World Health Organization (WHO) and the European Food Safety Authority (EFSA) have supported the TDS approach for several years, and have provided general methodological guidelines (EFSA, 2011a). Nevertheless, none of these documents have proposed a methodology to validate the appropriateness of this approach for assessing exposure to the different types of substances, given that not all substances can be evaluated through a TDS. A TDS follows three essential principles – i.e. (i) representativeness of the whole diet, (ii) pooling of foods, and (iii) food analyzed as consumed – and the applicability of the TDS approach for the different substance groups has to be considered with regard to these criteria.

The aim of this article is to propose a general method to validate the relevance of the TDS approach, and to apply it to different groups of substances, independently on the fact whether they have already been included in a TDS or not. Only chemical substances are dealt with in this work; biological agents have not been considered as their potential effects are mainly linked to acute exposure (TDS aim at assessing chronic and not acute exposures).

This study was carried out in the framework of the European project TDS-EXPOSURE and is a general analysis conducted for whole groups of substances. Some particular cases are sometimes identified, but in general the conclusions are given for groups of chemical agents and may not apply to some specific substances. For example, the TDS approach could be considered as generally not relevant for a group, even if some specific substances of the group may be studied in a TDS, taking particular caution with the method.

2. Materials and methods

2.1. Materials

As a first step, a list of chemicals to be considered in this work has been established based on different data sources: substances for which the EFSA published an opinion or received a mandate for assessment (74 substances or substance classes at the time of the assessment) (EFSA), substances for which the Joint FAO/WHO Expert Committee on Food Additives (JECFA) published an evaluation or a monography ($n = 2412$) (JECFA), substances included in the Codex Alimentarius ($n = 189$) and in the European rapid alert system for food and feed (RASFF) notifications ($n = 103$) (European Commission), and substances recorded since 2004 ($n > 30$) in the International Food Safety Authorities Network (INFOSAN). Due to the large number of existing chemicals it was not possible to be exhaustive, even though all of the most important substances occurring in food and specifically those recognized as being particularly critical in terms of their effects on human health were included. Most importantly, similar principles can be adopted for new sets of substances detected in future studies targeting new/emerging contaminants.

The substances selected were divided into eight main groups that can be present in the diet, based on the list of the EFSA/FAO/WHO guidance document (EFSA, 2011a): (i) natural components considered beneficial or essential (e.g. micronutrients such as vitamins, iron, iodine, and selenium), (ii) trace elements and contaminants from the environment (e.g. 'heavy metals', polychlorinated biphenyls and dioxins), (iii) chemical substances intentionally added to foods (e.g. preservatives and colours), (iv) chemical residues of substances being deliberately applied at other points in the food production chain (e.g. pesticides and veterinary drug resi-

dues), (v) naturally occurring contaminants (e.g. mycotoxins and alkaloids), (vi) contaminants formed during food processing (e.g. polycyclic aromatic hydrocarbons (PAHs), furan and acrylamide), (vii) contaminants transferred from food packaging or food contact materials (e.g. phthalates and bisphenol A), and (viii) other substances that have already been analyzed in one or more TDSs, including radionuclides (American TDS (FDA), Canadian TDS (Health Canada, 2009)), phytoestrogens (UK TDS (FSA), French TDS (ANSES)), or nitrosamines (Canadian TDS), and other chemicals that have never been studied in any TDS but that should be considered in this work, such as flavorings and nanomaterials.

2.2. Methods

The relevance of the use of the TDS approach for the different substance groups has to be considered with regard to three essential principles, i.e. a TDS (i) has to be representative of the whole diet, (ii) is based on pooling of foods, and (iii) involves food analyzed as consumed (EFSA, 2011a). The point is to check if these three principles would not exclude the evaluation of a family of substances. Four criteria have been considered for that purpose. The first criterion is that the substance has to be present in a significant part of the diet or predominantly present in specific and identified food groups. If the substance is present occasionally in a few foods, the TDS approach is not appropriate. The TDS sampling has to cover exposure from the whole diet, i.e. from all the potential food contributors to the exposure. The second criterion is related to the availability of a robust, ideally validated, analytical method to determine with adequately low LODs the chemical in potential food contributors to the exposure of the population. If it is not technically feasible to analyze the substance in all the potential food contributors or this is possible only in a limited portion of the food items (such as in fat for instance, if the substance can be present in the rest of the food), the TDS approach is not relevant. The third criterion consists in checking the dilution impact of pooling (effect of dilution and mixing of samples). If the concentration of the substance will be highly affected by pooling, e.g. because it occurs only in particular foods or is linked to specific geographical regions or is very volatile, so that it will become undetectable in pooled samples, then the TDS approach is not relevant. The fourth and last criterion is related to the possibility to mimic the impact of everyday preparation methods of food, in the household and other places and situations of daily life (e.g. restaurants, canteens, coffee shops) on the concentration of the substance. As foods are analyzed as consumed in a TDS, it is necessary to reproduce as precisely as possible everyday preparation methods, especially if the concentration is highly impacted by them (loss of volatiles, formation of heat-generated substances, pick-up of contaminants from cookware). If these methods are not known, are too variable or cannot be reproduced in the laboratory, the TDS approach may not be appropriate and/or may need supplementation by another study.

These four criteria are summarized in Table 1.

For each group of substances dealt with, the four criteria were applied to determine if the TDS approach was generally relevant or not, or relevant only in some specific cases.

3. Results and discussion

3.1. Natural components considered beneficial or essential

Major nutrients, in particular minerals, are present in a large part of the diet. The TDS approach is currently used in several countries to estimate the intake of nutrients, which are then compared to the recommended dietary allowances (Lombardi-Boccia et al., 2003; Lowik et al., 1994; Ockhuizen et al., 1991; Pennington and Schoen, 1996; Turrini and Lombardi-Boccia, 2002; van Dokkum et al., 1989) or to the upper limits (FSANZ, 2008).

Well-established methods are available to analyse nutrients in all food contributors, based on e.g. high-performance liquid chromatography (HPLC) for vitamins, inductively coupled plasma optical emission spectroscopy (ICP-OES) or flame atomic absorption spectrometry for macroelements. It is possible to analyse nutrients in all potential food contributors but special care should be taken of labile vitamins because endogenous content could dramatically decrease during the storage and the analytical procedure.

General considerations on the smoothing effect caused by pooling are applicable to nutrients. Pooling of samples in food groups would not highly dilute nutrients levels, but care should be taken in food group composition. When fortified foods and nutritional supplements are considered as individual food categories, pooling should be very carefully arranged, because the concentration of nutrients in this type of food is much higher than the one from

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