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Drinking water, diet, indoor air: Comparison of the contribution to environmental micropollutants exposure

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ABSTRACT

This study collated 254,441 analytical results from drinking water quality monitoring in order to compare levels of exposure of the French adult population from drinking water with that from total diet for 37 pesticides, 11 mineral elements, 11 polycyclic aromatic hydrocarbons (PAH), 6 non dioxin-like polychlorobiphenyls (NDL PCB), 5 ether polybromodiphenyl ethers (BDE), 2 perfluorinated compounds. It also compares levels of exposure from drinking water with that from inhalation of indoor air for 9 volatile organic compounds (VOC) and 3 phthalates. The vast majority of the water analysis results showed values below the limits of quantification and this comparison was primarily made on the basis of a highly pessimistic scenario consisting in considering the data below the limits of quantification as being equal to the limits of quantification. With this conservative scenario, it can be seen that tap water makes a minor but potentially non-negligible contribution for a few micropollutants, by comparison with diet and air. It also shows that exposure through drinking water remains below the toxicity reference values for these substances. Apart from a few extreme values reflecting exceptional local situations, the concentrations measured for the minority of positive samples (below the 95th percentile value) suggest a very low risk for human health. Lower limits of quantification would however be of use in better estimating the safety margin with regard to the toxicity reference values, in particular for BDE, PAH and NDL PCB.

Introduction

Humans are exposed to environmental micropollutants through a number of pathways, more specifically air, diet, water or contact with products and materials. Faced with the large number of micropollutants detected in all environmental compartments and the diversity of possible exposure pathways, it is important to identify the main pathways of transmission of these micropollutants to humans, so that the preventive measures to be taken to mitigate this exposure can be prioritised. Based on the surveillance data concerning the various compartments available today, the aim of this study was to identify the micropollutants which could be significantly transmitted by drinking water, as compared with other possible pathways. The aim was also to help to identify the micropollutants on which drinking water systems operators should concentrate their efforts in order to improve consumers' health protection.

In all developed countries, drinking water quality is closely monitored. Consequently, operators of the water services today have considerable quantities of surveillance results for contaminants, whether or not regulated, which can be used to assess exposure through drinking water.

Other than drinking water, few compartments are the subject of close monitoring for such a diversity of contaminants. To assess exposure through diet, the World Health Organisation (WHO) has since the early 1980s recommended a "total diet study" (TDS) approach (WHO, 1979). Practical advice for performing this

Abbreviations: ANSES, Agence Nationale de Sécurité Sanitaire de l'Alimentation, de l'Environnement et du Travail, *National Agency for Food, Environment and Occupational Safety*; ATSDR, Agency for Toxic Substances and Disease Registry; BDE, (poly) bromo diphenyl ether; EPA, Environmental Protection Agency; EFSA, European Food Safety Authority; FAO, Food and Agriculture Organization; FDA, Food and Drug Administration; JECFA, WHO/FAO Joint Expert Committee on Food Additives; JMPR, Joint FAO/WHO Meeting on Pesticide Residues; LOD, limit of detection; LOQ, limit of quantification; NDL PCB, non dioxin-like polychlorobiphenyl; OQAI, Observatoire de la Qualité de l'Air Intérieur, *Indoor Air Quality Observatory*; PAH, polycyclic aromatic hydrocarbon; RIVM, Rijksinstituut voor Volksgezondheid en Milieu, *National Institute for Public Health and the Environment*; TDS, total diet study; TEQ, toxic equivalent; TRV, toxicity reference value; VOC, volatile organic compound; WHO, World Health Organization.

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type of study was provided jointly by the WHO and the Food and Agriculture Organization (FAO) (WHO, 1985) and, further to these recommendations, numerous countries approved and followed this approach for assessing the exposure of their populations. Historically, the first countries to initiate this process were the United States as of 1961, through the Food and Drug Administration (FDA, online, http://www.fda.gov/food/foodscienceresearch/totaldietstudy/default.htm), Australia, which has just completed the 24th TDS (FSANZ, 2014) and Canada, with an initial study between 1969 and 1973 (Health Canada, online, http://www.hc-sc.gc.ca/fn-an/surveill/total-diet/index-eng.php). In Europe, the first study of this type was conducted in 1966 in the United Kingdom and it is regularly updated by the Food Standards Agency (FSA, 2009). Since the 1990s, more and more European countries have conducted total diet studies. This is the case with France, in which the first study was carried out from 2001 to 2004 (Afssa, 2004) with the aim of updating it every 5 years. The second French total diet study, called EAT 2, was carried out from 2006 to 2011 (Anses, 2011).

This study aims to be as complete as possible, with about 450 contaminants being looked for in more than 200 foodstuffs. The directly accessible results, as well as the methodology used, make it an ideal study for comparing contamination and exposure across the French population. It is based on the "INCA 2" individual and national study of food consumption (Afssa, 2007), which examined the dietary habits of more than 2500 French citizens. The contaminant concentrations measured in the foodstuffs are thus directly reconciled with the dietary habits and body weight of the participants in order to estimate the mean exposure of the population sample.

Although outdoor air quality has been the subject of numerous studies since the 1970s, indoor air quality is a more recent concern, even though populations in developed countries spend most of their time in enclosed spaces such as housing, offices, schools or transport systems (Zeghnoun et al., 2010). Since the French indoor air quality observatory (OQAI) was created in 2001, monitoring data on a certain number of micropollutants have been available to the public and enable exposure through indoor air to be estimated.

Given the available data on water, diet and indoor air monitoring in France, this study focused on the French adult population and on micropollutants the presence of which has been documented for at least two of these three exposure pathways.

Methodology

Exposure through diet

In this study, the results used are those of the second French total diet study EAT 2 (Anses, 2011). The chemical families considered include 445 potential food contaminants belonging to the following families: inorganic and mineral compounds, mycotoxins, phytoestrogens, polychlorodibenzo-*p*-dioxins and furanes (PCDD/F), dioxin-like (DL-PCB) and non-dioxin-like (NDL-PCB) polychlorobiphenyls, perfluorinated compounds, brominated flame retardants, pesticides, food additives, polycyclic aromatic hydrocarbons (PAH) and acrylamide.

The INCA 2 study was used to determine the foods analysed during EAT 2. Briefly, 9 lists of foodstuffs (1 national and 8 regional) were drawn up for a total of 212 types of foodstuffs. These products were collected from the whole of France during two different seasons (spring/summer and autumn/winter). They were then prepared "as consumed" (peeling, washing, cooking, etc.). Only the inorganic compounds were looked for in all the foodstuffs studied, while the other compounds were only looked for in the foodstuffs most likely to be contaminated by the pollutant in question.

Dietary exposure of the population to each contaminant of interest was individually calculated using the following equation (Anses, 2011).

$$E_{i,j} = \frac{\sum_{k=1}^{n} C_{i,k} \times T_{k,j}}{PC_i} \quad (1)$$

where $E_{i,j}$ exposure of individual i to contaminant j (ng d^{-1} kg^{-1}), n number of foodstuffs in the diet, $C_{i,k}$ consumption of foodstuff k by individual i (g d^{-1}), $T_{k,j}$ content of contaminant j in foodstuff k (ng g^{-1}), PC_i body weight of individual i (kg).

This formula was applied individually to all the participants in the INCA 2 study, using their declared foodstuffs consumption and their measured weight. The measurement results for substances below the detection or quantification limit were considered by the authors in two ways (Anses, 2011):

If less than 60% of the measurement results were below the limit of detection (LOD), the concentrations below the LOD were considered to be equal to 1/2 LOD and the concentrations lower than the limit of quantification (LOQ) equal to 1/2 LOQ (mean hypothesis),

If 60% or more of the measurement results were below the LOD, two hypotheses were considered: a low hypothesis with concentrations below the LOD equal to 0 and the concentrations below the LOQ equal to the LOD, and a high hypothesis with the concentrations below the LOD considered to be equal to the LOD and the concentrations below the LOQ considered to be equal to the LOQ.

The exposure calculated was then interpreted by comparison with toxicity reference values (TRV) established by national (ANSES, ATSDR, RIVM, USEPA), European (EFSA) or international scientific agencies and workgroups (JECFA, JMPR, WHO).

The dietary exposure evaluated by the EAT 2 study included drinking water. However, the exposure data due specifically to drinking water were not published.

Exposure through drinking water

In order to evaluate the exposure specifically through drinking water, the contaminant concentrations used were those measured during the monitoring and quality control of the waters produced and distributed by Lyonnaise des Eaux in France between 2009 and 2012. For a few contaminants (cadmium, VOC and BDE), older results obtained between 2002 and 2012 were used, in order to increase the total number of data. The company operates about 1400 production units in France and supplies drinking water to about 12 million people, representing almost 20% of the French population. Approximately 65% of the volumes produced (and consequently of the samples collected for quality monitoring) originate from ground waters, and the remaining 35% from surface waters. The treatment processes involved range from a simple chlorination in the case of well protected ground waters, to full conventional treatment for surface waters, including coagulation (with aluminium salts in most cases), sedimentation, rapid sand filtration, ozonation, granular activated carbon filtration, and chlorination. The treatment can also include membrane ultra-filtration and UV disinfection in some cases. Samples for quality monitoring in production are taken at the treatment plant outlet, and samples for quality monitoring in distribution are taken in storage tanks and at consumers' taps. Samples in production and distribution are collected after a minimal flushing time of 30 s, according to standards ISO 5667-3 (2003) and ISO 5667-5 (2006). The analytical methods applied for the micropollutants considered in this study are summarized in Table 1. When LOQs changed during the study period, the results were interpreted with regard to the highest LOQ observed over the study period. Eq. (1) was applied to evaluate the exposure of the adult population through drinking water. As an exposure estimate for each individual was impossible, a daily

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