



# Which compounds contribute most to elevated airborne exposure and corresponding health risks in the Western Balkans?

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## ABSTRACT

A majority of ongoing monitoring of persistent organic pollutants (POPs) is currently focused on chemicals emphasized in the Stockholm Convention. Quantitative detection of other substances (especially those with numerous anthropogenic sources such as polyaromatic hydrocarbons (PAHs)) is, however, also needed since their concentrations are usually several orders of magnitude higher. A goal of this study was to determine how various groups of compounds contribute to total human health risks at the variety of sampling sites in the region of Western Balkan. Distribution of the risks between the gas and particulate phases was also addressed. Results showed that inhalation exposure to organochlorine pesticides (OCPs) does not represent a significant risk to humans, while polychlorinated biphenyls (PCBs) re-volatilized to the atmosphere from contaminated soils and buildings can pose a problem. PCB evaporation from primary sources (currently used PCB-filled transformers or non-adequate storage facilities) generally resulted in much higher atmospheric concentrations than evaporation from the secondary sources (soils at the sites of war destructions). A majority of the human health risks at the urban sites were associated with PAHs. Between 83 and 94% of the cumulative risk at such sites was assigned to chemicals sorbed to particles, and out of it, PAHs were responsible for 99%.

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## 1. Introduction

A potential impact of polluted air on human health has been a subject of numerous investigations recently. It has been suggested that air pollution is likely to increase mortality and hospital admissions (Brunekreef and Holgate, 2002). The inhalation exposure can result in a range of effects from breathing difficulties to development of a lung cancer (Boffetta and Nyberg, 2003; Vineis and Husgafvel-Pursiainen, 2005). In addition to respiratory tract, vital functions of other organs can be affected as well (Cohen et al., 2005; Kunzli and Tager, 2005). A quantification of such harmful effects is complicated by the fact that ambient air is a complex mixture of components with variable chemical composition, physicochemical properties, persistence in the environment, long-range transport potential, toxicity and carcinogenicity.

Several regulations on production, marketing, application and disposal of persistent toxic substances have been introduced in recent years but many of these compounds persist in the environment at significant levels long after they have been banned. To evaluate the effectiveness of recently introduced regulations on production, marketing, application and disposal of persistent toxic substances; reliable air monitoring programs are needed worldwide. As many of these compounds persist in the environment long after they have

been banned, such programs should provide information on present concentrations and their long-term trends (Middleton, 1997). Ambient air concentrations derived from the regular monitoring of POPs are also a source of data for evaluation of the long-term population exposure and related effects, in particular for pollutants for which ambient air represents a dominating exposure pathway.

A majority of ongoing monitoring of persistent organic pollutants (POPs) is currently focused on chemicals emphasized in the Stockholm Convention (UNEP, 2001) which regulates emissions of compounds such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and furans (PCDDs/Fs), and organochlorine pesticides (OCPs). Quantitative detection of other substances, (especially those with numerous anthropogenic sources such as PAHs) is, however, also needed since their concentrations are usually several orders of magnitude higher (PCDDs/Fs:  $\text{pg m}^{-3}$  or less, PCBs and OCPs: tens to hundreds of  $\text{pg m}^{-3}$ , PAHs up to hundreds of  $\text{ng m}^{-3}$ ) (Menichini et al., 2007). Even though reduction of the human exposure to POPs in ambient air has been a primary focus of the public health policy, the lack of available data often prevents the authorities from the adequate actions.

Countries of the former Yugoslavia represent a European region with very limited information on the levels of atmospheric pollution. As many industrial sites and energy installations were damaged in the bomb attacks and missile strikes during the Balkan war conflicts in the late 1990s, and large amounts of hazardous substances were released into the environment (Picer and Holoubek, 2003), contamination of

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the former Yugoslavia has been a subject of numerous investigations in the last decade (Rapsomanikis et al., 2002; Picer and Holoubek, 2003; Klánová et al., 2007a,b; Ruzickova et al., 2008). The scientific attention was focused mostly on determination of the PCB concentrations in soil and air, while other classes of pollutants were largely overlooked. A first study investigating toxicity of the air samples from this region, however, showed that toxic effects were much stronger (Skarek et al., 2007) in the air samples from industrial sites contaminated with PAHs than in the samples from PCB hotspots.

A contribution of various classes of toxic compounds (PCBs, OCPs, PAHs), both particle-bound and gas phase-associated, to total human health risks was assessed in this study. Large number of ambient air samples from background, urban and industrial sites including hot spots in Croatia, Serbia, Bosnia and Herzegovina was collected. Ambient air concentrations of selected pollutants and related human health risks associated with the inhalation exposure were determined for all samples.

## 2. Materials and methods

### 2.1. Sampling locations

A total number of 127 ambient air samples were collected in the countries of former Yugoslavia (Croatia, Serbia, Bosnia and Herzegovina) in the early summers (May–June) of 2003 and 2004 under comparable meteorological conditions at all sites (median temperature 19°C). In 2003, ten high volume samples were collected from each of five sampling sites in Croatia. Four samplers were positioned in the city of Zadar (damaged transformer station, industrial zone, historical center, meteorological station). Background sampler was 150 km north on mountain Velebit at the Zavizan location. Similarly, five samples were taken from each of five sampling sites in Sarajevo area (industrial zones, residential areas, background site) and another five sampling sites in Tuzla region in Bosnia and Herzegovina (service and storage place for damaged capacitors, transformer station, salt mine, residential area, background site). Ivan Sedlo meteorological station served as a background site for Bosnia and Herzegovina. In Serbia, Kragujevac (Zastava factory, university), Pancevo (Petrochimika, refinery, center) and Novi Sad (refinery, residential part, center) were the cities of interest, each providing three sampling sites and three high volume samples from each site.

### 2.2. High volume air sampling

The high volume air samplers PS-1 (Graseby-Andersen, USA, flow: 20–25 m<sup>3</sup> h<sup>-1</sup>, volume: 250–300 m<sup>3</sup> per 24 h) and two types of adsorbents were used: a Whatmann quartz filter (fraction <50 μm) for a collection of particles, and a polyurethane foam filter (Gumotex Břeclav, density 0.03 g m<sup>-3</sup>) for a gaseous phase sampling. A sampling duration was 24 h. All filters were cleaned before the campaign: PUF filters were extracted with acetone and dichloromethane in a Soxtec extractor, quartz filters were heated to 450 °C.

### 2.3. Passive air sampling

To assess the seasonal variability of the air concentrations of investigated POPs, passive air sampling technique has been employed at all sampling sites for the next six months following the active air sampling. Passive air samplers consisting of the polyurethane foam disks (15 cm diameter, 1.5 cm thick, density 0.030 g cm<sup>-3</sup>, type N 3038; Gumotex Břeclav, Czech Republic) housed in protective chambers (Shoeb and Harner, 2002; Klanova et al., 2006) were employed in this study. Sampling chambers were washed and solvent-rinsed with acetone prior to installation. All PUF disks were prewashed, cleaned (8 h Soxhlet extraction in acetone and 8 h in dichloromethane), wrapped in two layers of aluminum foil, placed

into zip-lock polyethylene bags and kept in a freezer prior to deployment. Exposed PUF disks were wrapped in two layers of aluminum foil, labeled, placed into zip-lock polyethylene bags and transported in cooler at 5 °C to the laboratory where they were stored at –18 °C until analysis. Field blanks were obtained by installing and removing the PUF disks at all sampling sites.

### 2.4. Sample analysis

All samples were extracted with dichloromethane in a Büchi System B-811 automatic extractor. One laboratory blank and one reference material were analyzed with each set of samples. Surrogate recovery standards (D8-naphthalene, D10-phenanthrene, D12-perylene for PAHs analysis, PCB 30 and PCB 185 for PCBs analysis) were spiked on each filter prior to extraction. Terfenyl and PCB 121 were used as internal standards for PAHs and PCBs analyses, respectively. Volume was reduced after extraction under a gentle nitrogen stream at ambient temperature, and fractionation achieved on a silica gel column (30 cm length, 1 cm i.d.); a sulphuric acid modified silica gel column was used for PCB/OCP samples. Samples were analyzed a GC-MS instrument (HP 6890–HP 5975) supplied with a J&W Scientific fused silica column DB-5MS for PCBs (PCB 28, PCB 52, PCB 101, PCB 118, PCB 153, PCB 138, PCB 180), OCPs (α-HCH, β-HCH, γ-HCH, δ-HCH, p,p'-DDE, p,p'-DDD, p,p'-DDT) and 16 US EPA PAHs. Analytical details and Quality Assurance/Quality Control measures have been published previously (Klánová et al., 2007a,b).

### 2.5. Risk assessment

According to EPA (EPA, 1998), a human exposure depends on a Chronic Daily Intake (CDI) of every single contaminant inhaled by the receptor. The CDI value (mg kg<sup>-1</sup> day<sup>-1</sup>) can be derived from Eq. (1) (EPA, 1992; EPA, 1996):

$$CDI = CA \cdot IF, \quad (1)$$

where CA is a compound concentration (mg m<sup>-3</sup>) and IF is an Intake Factor (m<sup>-3</sup> kg<sup>-1</sup> day<sup>-1</sup>).

Intake Factor is derived from Eq. (2):

$$IF = \frac{IR \cdot A \cdot EF \cdot ED \cdot ET}{BW \cdot AT}, \quad (2)$$

where IR-A (Inhalation Rate) is a breathing rate (m<sup>3</sup> h<sup>-1</sup>), EF (Exposure Frequency) is a number of exposures per year, ED (Exposure Duration) is a duration of exposure in years, ET (Exposure Time) is a number of hours per exposure, BW (Body Weight) is a default weight of the receptor body (kg), and AT (Averaging Time) is an average exposure extent over a lifetime (35,500 days for carcinogenic exposure). Appropriate default exposure parameters were obtained from EPA (EPA, 1998) [IR-A = 20 m<sup>3</sup>/day; EF = 365 days; ED = 70 years; ET = 24 h/day; BW = 70 kg]. CDI for carcinogenic substances is called Life Averaged Daily Dose (LADD).

Human health risk related to contaminated air depends on the extent of exposure as well as on the toxic effects of chemicals. The chemical-specific risks were calculated from the Life Averaged Daily Dose and the Slope Factor (SF) (1/mg kg<sup>-1</sup> day<sup>-1</sup>) using the linear low-dose cancer risk equation (Eq. (3)):

$$Risk = LADD \cdot SF \quad (3)$$

A Slope Factor is a plausible upper-bound estimate of probability of the response per unit chemical intake over the lifetime. It is used to estimate an upper-bound probability of the individual developing a cancer as a result of the lifetime exposure to certain level of potential carcinogen. Cancer potency factors for chemicals of concern were obtained from EPA, OEHHA (OEHHA, 2002; EPA, 2003). Cancer

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