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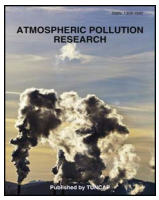

Urban environment as a factor modulating metals deposition in the respiratory track and associated cancer risk

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ABSTRACT

The aim of this study was to compare the inhalation cancer risk posed by PM-bound As, Cd, Pb, Ni and Cr(VI) concentrations in typical urban environment and to differentiate this risk between PM size fractions and population age-groups. Additionally inhalation deposition metrics were used to characterize risks posed by PM of highway (H)-, street canyon (SC)- and residential (RA) origin. Aerosol samples were collected in the cities of Zabrze and Katowice (Poland) using Dekati cascade impactors. Regardless of the location metals were primarily associated with particles less than 1 μm. For sites, where the PM size distribution was bi-modal (SC in Zabrze and Katowice and RA in Zabrze) and the maxima of metal mass distribution occurred in both accumulation and coarse modes, the metal was predominantly deposited in head airways region. Sites characterized by an unimodal distribution (RA in Katowice, H in Katowice and SC in Zabrze) were found to favor pulmonary deposition. The overall mass deposition of metals in the respiratory tract in residential; highway and street canyon environments were: 0.51; 0.54 and 0.61, respectively. Street canyon concentrations were therefore most hazardous. Depending on the specific compartment of the respiratory tract, average metal mass deposition amounts varied from 0.05 to 0.53 of its ambient concentration. ILCR values were up to 10E-03 for adults versus 10E-05 for infants. Adults were mostly exposed near street canyons, while infants and children were mostly susceptible near residential and highway environments.

1. Introduction

Epidemiological evidence suggests, that the carcinogenic potential of particulate matter (PM) is partially attributable to carcinogenic metals that it contains (Gutiérrez-Castillo et al., 2006; Huang et al., 2013). Results from exposure studies in the workplace such as smelters or cooking plants, indicate elevated mortality or incidence of lung cancer among workers exposed to welding fumes or particles containing metals (IARC, 1993; Wild et al., 2000; Goldberg and Goldberg, 2003; Beveridge et al., 2010; 't Mannetje et al., 2011). Atmospheric concentrations of metals are generally substantially lower than concentrations found in the workplace. In some regions of the world or in close proximity to industrial sources, ambient concentrations may be very high, in some cases exceeding the National Air Quality Standards by many times (Rogula-Kozłowska et al., 2013, 2015; Mondol et al., 2014; Islam et al., 2015; Li et al., 2016). Such high concentrations typically occur in urban areas, where combustion of fossil fuels for heat generation together with traffic and industrial emissions negatively impact air quality (Rogula-Kozłowska et al., 2014, 2015, 2016b). The contribution of sources varies not only between cities but also within urban environment (Rogula-Kozłowska et al., 2015, 2016b; Olszowski, 2016, 2017). In fact, each city location (e.g. roads, pavement, settlements and parks) is characterized by individual emission profiles, and therefore contributes differently to PM and PM-bound metals related exposures. Estimates of the distribution of exposures is critical to understanding how differences in PM composition contribute to adverse health effects, such as respiratory functions and lung cancer. Information concerning PM-bound metals concentrations and their seasonal patterns alone is insufficient to quantify small differences in exposure levels (Widziewicz and Loska, 2016). This is especially true in areas, where PM profiles are obscured by municipal emissions, primarily from hard coal combustion (observed especially during cold weather), such as in Upper Silesia (Poland) (Rogula-Kozłowska et al., 2013, 2015). In these cases additional information concerning PM size-fractionation and size-dependent deposition in the respiratory tract could help give insight into exposure routes between nearby microenvironments. Recent research and epidemiological studies indicate, that elevated levels of fine metal-bearing particles are associated with more fatalities, than

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coarse fractions (Utsunomiya et al., 2004; Valavandis et al., 2008). This suggests that areas, where metals are present in the fine PM (those easily migrating into the lungs) pose greater health hazard than locations where PM is dominated by coarse PM (earth crust material, wind-blown dust, dust from resuspension, soot, etc.). It can therefore be concluded that the inhalation exposure to airborne carcinogenic metals will vary depending on their ambient concentrations, distribution within space, site-specific emission profiles and metals concentrations in the air.

Since city resident are inevitably exposed to different concentrations of PM-bound metals, it is crucial to study, which location within the city pose the greatest health impacts. Such information could provide some tips concerning healthier options for city residents. There are no published data of metal exposure of Poles exposed in their microenvironments during normally daily activities. Therefore as a scope of this work we decide to look for such exposure differences, interestingly focusing on a small urban area, located in a European hotspot region, where annual concentrations of PM-bound metals are even higher than in biggest Asian metropolises (Rogula-Kozłowska, 2015).

To determine the potential role of different urban microenvironments on PM-bound metal exposures and associated inhalation cancer risks, the present study utilized a novel integrated methodology including PM-size fractionation and deposition modelling to apportion exposures to street canyon, highway and residential microenvironments. By incorporating additional environmental information, we were able to differentiate exposures between different age groups, as well as between near-traffic and urban background sites.

2. Methodology

2.1. Characteristic of sampling sites and measurement campaign

Measurements were carried out in Silesia Province – (śląskie; southern Poland; Fig. 1), a region characterized by the worst air quality in the entire country and known European hotspot (e.g. Kiesewetter et al., 2015). PM samples were collected at five sites, differing significantly in amounts and structures of PM emissions.

Three sites were located in Katowice city (first one in the geographic heart of a city – commonly the city center near the two busy crossroads and the largest roundabout in Katowice (1); second one in the Katowice suburbs – namely residential site (2) and the third one in the close proximity to the A4 highway (3). Two remaining sites were located in Zabrze city and included: residential area (4) with a highly developed building and typical street canyon in the city center (5).

A detailed description of the sampling sites is presented in Table 1. For collection of PM samples, a thirteen-stage low pressure impactor (DLPI website, Dekati Ltd.; Kangasala, Finland, flow rate 30 l/min; pump: Sogevac SV/25) was used. Seventy eight thirteen-fold PM samples were taken during the sampling period. DLPI splits PM into 13 PM fractions: $PM_{0.03–0.06}$, $PM_{0.06–0.108}$, $PM_{0.108–0.17}$, $PM_{0.17–0.26}$, $PM_{0.26–0.4}$, $PM_{0.4–0.65}$, $PM_{0.65–1.0}$, $PM_{1.0–1.6}$, $PM_{1.6–2.5}$, $PM_{2.5–4.4}$, $PM_{4.4–6.8}$, $PM_{6.8–10.0}$, and $PM_{10.0–100}$, where PM_{x-y} denotes the PM fraction whose particles have their D_p in the interval [x,y]. $PM_{0.03–0.06}$ (1-st stage) and $PM_{10.0–100}$ (13-th stage) represented exit and inlet particles (Hinds, 1998). Particular sample collection lasted from 69 to 173 h and was conducted during non-heating periods (i.e. spring and summer). A novel approach was used to ensure winter concentration data was not masked by emissions from coal combustion. During the winter, both atmospheric aerosol loading and its composition in Silesia, is dominated by carbonaceous municipal emissions, which has been identified as a main contributor to PM and PM-bound metal pollution (Pastuszka et al., 2010; Rogula-Kozłowska et al., 2014). Normally the average, annual concentrations of As, Cd, Ni, and Pb in Poland are characterized by little variation (Supplemental Information; Table S1). Treating exposure as a result of annual averages, one can mistakenly conclude that there are small variations in risk, not only at the city scale (e.g. Zabrze and Katowice) but also throughout the whole country. By using our approach it is possible to differentiate risk levels even in a relatively small urban area.

Due to chemical purity and low metal contents nylon membrane filters (0.2 μm, ø25 mm, Whatman) were used as collection substrates. PM mass concentrations were calculated by weighing filters before and after sampling using a MYA 5.3Y.F micro balance (RADWAG; Radom, PL), with a 1 μg resolution. Substrates were conditioned for 48 h in the weighing room (relative air humidity 45% ± 5%, air temperature 20 ± 2 °C) prior to weighing. After weighing, exposed substrates were stored in a freezer until analysis.

2.2. Instrumental analysis

The PM samples (1014 in total) were analysed for their elemental content (Cr, Ni, As, Cd and Pb) using a PANalytical Epsilon 5, EDXRF analyser with a side-window, liquid-cooled, 100 keV, Gd anode x-ray tube, and secondary targets, for: Al, Ti, Fe, Ge, Zr, Mo, Ag, Ce_2O_3, and Al_2O_3. Calibration was performed by measuring thin-layer single-elements standards (Micromatter; Vancouver, BC, CA). Analysis, included the entire program of X-ray tube settings (25 keV and 25 mA for Al, 40 keV and 15 mA for Ti, 40 keV and 15 mA for Fe, 75 keV and 8 mA for Ge, 100 keV and 6 mA for Zr, 100 keV and 6 mA for Al_2O_3) lasted 2400 s. The analytical procedure was verified weekly with a reference material (SRM2873, NIST). Element recovery was between 87% (As) and 116% (Pb) of the certified value. Three blanks (non-exposed Nylon membrane filters) were used to determine the detection limits for the procedure. Each blank underwent the entire EDXRF procedure repeated 30 times. Detection limit was calculated as the standard deviation from the 90 results obtained for each element. Detection limits were between 0.4 (Pb) and 7.1 (As) ng cm^{-2}. Measurement results including PM_1, $PM_{2.5}$ and PM_{10} mass concentrations and PM_1-, $PM_{2.5}$- and PM_{10}- bound element concentrations at selected sampling sites are presented in Fig. 2.

Typical concentrations of As, Cd, Ni and Pb in Poland obtained within the Polish State Environmental Monitoring Program are in Supplemental Information (Tables S1 and S2). The summary gives annual concentrations of As, Cd, Pb, and Ni at urban background sites in the Silesia region, as well as at rural background sites in Poland, over the past twelve years. However this summary lacks levels of those elements at traffic sites. Only two Silesia monitoring stations collect data at traffic stations (SlCzestCzes_arkr1 and SlKatowKato_aleja). None of them however, monitor concentration of interesting metals.

2.3. Respiratory tract deposition model

The deposition of PM-bound elements in the human respiratory track was calculated using the Multiple-Path Particle Dosimetry model (MPPD V2.11, ARA Inc.) (RIVA Report). This model is commonly used for estimating human airway particle dosimetry in three anatomically distinct compartments of the respiratory track: head airways (H), trachea -bronchiolar (TB) and pulmonary alveolar (P). A detailed description of this model can be find in the report published by RIVM, National Institute for Public Health and the Environment (RIVM, 2002). To model metals deposition, MPPD software requires particle properties as input parameters, including mass median aerodynamic diameter (MMAD) and geometric standard deviation (GSD), which were calculated from the sampled PM size ranges. For modelling purposes it was assumed that the mass percentage of each element, which is inhaled and deposited within the respiratory tract is equal to the inhalability of its individual metal-bearing particles. This assumption was necessary because the MPPD model is based on results from PM deposition studies and therefore does not allow users to specify extent to which compounds such as metals or PAHs are deposited and cleared after they are inhaled. Calculations were started by checking the mass size distribution of PM mass and its modality. This information was than used to