Science of the Total Environment 544 (2016) 391–399

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Evaluation of atmospheric inputs as possible sources of antimony in pregnant women from urban areas

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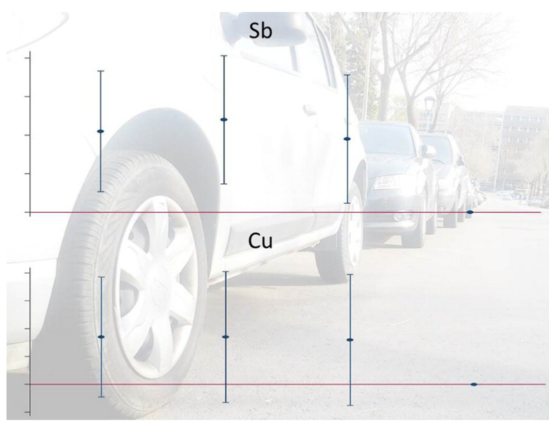
HIGHLIGHTS

- Atmospheric inputs are the main source of antimony for pregnant women in urban areas.
- Traffic pollution is a potential source of Sb for pregnant women in urban areas.
- There is an association between higher urine Sb and maternal physical activity.
- The association between atmospheric inputs and Cu urine levels is not significant.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 30 July 2015
Received in revised form 13 November 2015
Accepted 20 November 2015
Available online xxxx

Editor: D. Barcelo

Keywords:
Antimony
Copper
Pregnant women
Atmospheric pollution
Urine studies
Urban areas
Physical activity
Seasonal changes

ABSTRACT

Antimony and copper are common components of brake linings. The occurrence of these two metals in urban atmospheric aerosols has been related to vehicular use. Urine samples (n = 466) taken during the 32nd week of pregnancy were analyzed for Sb and Cu in pregnant women from an urban area (Sabadell, Catalonia, Spain). The geometric mean levels were 0.28 and 13 μg/g creatinine, respectively. Positive significant associations between urine concentrations of Sb and seasonality, intensity of physical exercise, working activities and traffic intensity at their home streets were observed. Cu showed the same trends but without statistical significance. In both cases, the estimated dietary ingestion of these two metals was larger than the inhalation inputs but the difference was much higher for Cu than for Sb. While Sb has no dietary role, Cu is an essential element which is also incorporated into humans through diet. The results suggest that inhalation of atmospheric particles may also constitute a source of Sb in pregnant women and general population of urban areas.

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http://dx.doi.org/10.1016/j.scitotenv.2015.11.095

1. Introduction

Atmospheric particles in urban areas have been linked to several health outcomes such as oxidative stress, inflammation, chronic obstructive pulmonar disease and cardiovascular or cerebrovascular stroke (Perez et al., 2009; Pope and Dockery, 2006). Significant correlations between daily mortality and ambient air particulate matter (PM) have been identified in Barcelona (Ostro et al., 2011). However, urban particles constitute very complex mixtures. Insight on the origin of these deleterious health effects and possible remediation actions depend on the association of specific properties such as size, chemical composition and others to specific toxic outcomes.

Sb is a toxic metalloid that is present in the diet at low concentrations (Arnich et al., 2012). The intestinal absorption of this element in humans is 5–20% (Lauwers et al., 1990). Its high volatility involves high affinity for atmospheric PM which constitutes a potential pollution source of this compound by particle inhalation (Belzile et al., 2012). Few studies on concentrations of Sb in humans are available (Filella et al., 2013a, 2013b) and very limited information is found on prenatal and children exposure but its presence in amniotic fluid has been observed (Caserta et al., 2011).

Cu is an essential metal that is necessary for the function of some enzymes such as ceruloplasmin or cytochrome c oxydase. It is present in a wide variety of foods (Mason, 1979). Besides diet and the gastrointestinal system, it may also be incorporated through respiration (Wiseman and Zereini, 2014). To date, industrial activity is the main source of this metal to the environment but vehicular traffic has also become a potential source because of its current use in brake linings (Amato et al., 2009). This metal is also known for its toxicity at high concentrations, children are more susceptible to deleterious effects than adults (Mason, 1979).

Sb and Cu are present in urban particulate material (Amato et al., 2011). Inhalation of PM may be a source of these metals for human populations. Urine is an adequate matrix for heavy metals biomonitoring (Fort et al., 2014) and can be collected without invasive methods. The concentrations of Sb and Cu in women at any pregnancy stage provide representative results for the whole pregnancy period (Fort et al., 2014). In the present study urban atmospheric pollution is evaluated as potential source of prenatal exposure to these metals.

2. Materials and methods

2.1. Urine samples

A cohort of 657 pregnant women were recruited between 2004 and 2006 in their 12th week medical visit (Primary Care Center II Sant Fèlix of Sabadell, Catalonia) within the INMA research network (INfancia y Medio Ambiente — Childhood and environment) (Guxens et al., 2012). Recruitment conditions were: residence in Sabadell, age higher than 16 years, single pregnancy, volunteering for the program and scheduled birth at the Hospitals of Sabadell or Terrassa (a nearby city). Women suffering from chronic diseases, having communication impairment or pregnancy from assisted reproduction were excluded. After obtaining the consent from the admitted women, questionnaires were administered by trained interviewers in the 12th and 32nd weeks of pregnancy. Dietary information was obtained by food frequency questionnaires obtained at both periods.

Pregnant women from this cohort provided a urine sample during the 32nd week of pregnancy ($n = 466$) which was collected in 100 mL polypropylene containers. The samples were stored in polyethylene tubes at −20 °C until analysis. This study was approved by the Research Ethics Committee of the CREAL. All information on participants was coded to maintain confidentiality.

2.2. Chemical analysis

Aliquots (3 mL) of each urine sample ($n = 466$) were introduced in Teflon vessels together with 3 mL of Instra-Analyzed 65% HNO3 (J.T. Baker, Germany) and 1.5 mL of Instra-Analyzed 30% H_2O_2 (Baker). The vessels were closed and heated at 90 °C in an oven overnight. After cooling, the vessels were opened and placed on a plate heated at 250 °C to evaporate the nitric acid. The resulting solid samples were dissolved with 3 mL of 4% HNO3 and stored in 7 mL plastic bottles which were subsequently kept in a refrigerator until instrumental analysis (Castillo et al., 2008; Krachler et al., 1998). This digestion protocol was validated by processing a Bio-Rad Level 1 urine reference sample (Lyphochek Urine Metals Control 1–69,131; Marnes-la-Coquette, France) that contains metal concentrations close to those of the urine from our studied population. The resulting inter-assay relative standard deviation coefficients were 17% and 4% for Cu and Sb, respectively. Before analysis, an internal standard of 10 ppb of In was introduced and, depending on sample density, samples were diluted with MilliQ water to 30 mL or 60 mL to avoid non-spectral interference. Instrumental analysis was performed by a Q-ICP-MS X-SERIES II instrument (Thermo Fisher SCIENTIFIC). One MilliQ water blank was processed together with each batch of samples to control for possible contamination. Instrumental limit of detection was 0.2 ng/mL attending to the most reliable lowest calibration point. A concentration of 0.1 ng/mL was established for samples under limit of detection for statistical purposes.

All wet-lab material was thoroughly cleaned by soaking in 10% nitric acid for 24 h, which was followed by three rinses of Milli Q water. The Teflon vessels were cleaned after every use by rinsing with 10% nitric acid (three times), then heating in the oven at 90 °C overnight, and finally rinsing with a high amount of Milli Q water.

Creatinine was determined by the Jaffé method (kinetic with target measurement, compensated method) with Beckman Coulter© reactive in AU5400 (IZASA®).

2.3. Statistical analysis

Arithmetic and geometric means, standard deviations (SD), medians, percentiles, minimum and maximum values of Sb and Cu in the studied population groups were calculated for descriptive statistics. Normality was checked by the Kolmogorov-Smirnov test.

Pregnant women included in the study answered to questionnaires regarding lifestyle and environmental exposures as well as food frequency questionnaires, conducted by trained interviewers. Exposure to vehicular traffic in environmental questionnaires was classified in four groups of intensity at home street, namely rare, moderate, frequent and heavy.

Sampling season and physical activity were also considered for their potential influence on atmospheric pollution intake. The former was assigned attending to sampling date and annual distribution of seasons, while the second referred to self-reported total physical activity, classified in three categories, namely sedentary or little active, moderately active and quite or very active. Type of maternal and paternal occupation (manufacturers, non-manufacturers), height of the housing and working time during pregnancy were also considered. The Sb and Cu levels between different categories were assessed by univariate linear regression modeling of the log-transformed concentrations of each metal and the categorical variables. Maternal age, pre-conception BMI, parity (in three categories, namely primiparous, one previous children and two or more previous children), social class, cotinine in urine and weekly consumption of the main groups of dietary items included in the food frequency questionnaires were also tested, as these could be associated with metal concentrations. Finally, stepwise regression was performed for the selection of variables ($p < 0.20$) included in multivariate linear regression. Interactions between car traffic exposure and season, physical activity, working time during pregnancy and height of the housing were assessed.