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# Small scale spatial gradients of outdoor and indoor benzene in proximity of an integrated steel plant



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#### HIGHLIGHTS

#### GRAPHICAL ABSTRACT

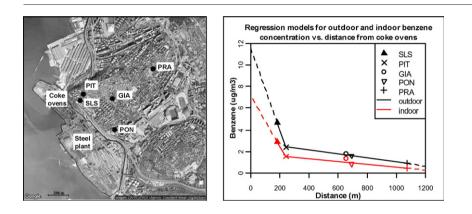
- Outdoor and indoor benzene data were collected in five dwellings close to a steel plant.
- The three closest sites exceeded the WHO reference level for benzene  $(1.7 \,\mu\text{g/m}^3)$ .
- Indoor benzene concentration was above 2 µg/m<sup>3</sup> in the dwellings closest to the works.
- The coke ovens were the main benzene source identified by wind regime and B/T ratio.
- A regression model of indoor vs. outdoor benzene concentration has been calculated.

#### A R T I C L E I N F O

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#### ABSTRACT

Benzene is known as a human carcinogen, whose annual mean concentration exceeded the EU limit value (5 µg/ m<sup>3</sup>) only in very few locations in Europe during 2012. Nevertheless 10% to 12% of the EU-28 urban population was still exposed to benzene concentrations above the WHO reference level of 1.7 µg/m<sup>3</sup>. WHO recommended a wise choice of monitoring stations positioning in proximity of "hot spots" to define and assess the representativeness of each site paying attention to micro-scale conditions. In this context benzene and other VOCs of health concern (toluene, ethylbenzene, xylenes) concentrations have been investigated, with weekly passive sampling for one year, both in outdoor and indoor air in inhabited buildings in close proximity (180 m far up to 1100 m) of an integrated steel plant in NE of Italy. Even though the outdoor mean annual benzene concentration was below the EU limit in every site, in the site closest to the works the benzene concentration was above  $5 \,\mu\text{g/m}^3$  in 14 weeks. These events were related to a benzene over toluene ratio above one, which is diagnostic for the presence of an industrial source, and to meteorological factors. These information pointed at the identification of the coke ovens of the plant as the dominant outdoor source of benzene. Benzene gradients with the increasing distance from coke ovens have been found for both outdoor and indoor air. Linear models linking outdoor to indoor benzene concentrations have been then identified, allowing to estimate indoor exposure from ambient air benzene data. In the considered period, a narrow area of about 250 m appeared impacted at a higher degree than the other sites both considering outdoor and indoor air. Passive BTEX sampling permits to collect information on both ambient air and daily life settings, allowing to assemble a valuable data support for further environmental cost-benefit analyses. © 2016 Elsevier B.V. All rights reserved.

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#### 1. Introduction

Benzene is a known toxic carcinogen (Snyder, 2012; Smith, 2010; IARC International Agency for Research on Cancer, 1989) and epigenetic modifications were related to benzene low dose exposures (Fustinoni et al., 2013).

Environmental exposure has been considered in general less relevant than cigarette smoke and occupational exposure (Bahadar et al., 2014; Manno, 2013; Lim et al., 2012; Wallace, 1996); nevertheless manifold non-cancer adverse health effects such as asthma, bronchitis, cardiovascular diseases, pollen and food sensitization, reproduction and development alterations are associated with a range of ambient exposure of  $1.01-24.8 \ \mu g/m^3$  (Bolden et al., 2015).

Accordingly to the recent European Environment Agency report on air quality in Europe (EEA, European Environment Agency, 2014) which refers to the three-year period 2010–2012, exceedances of the limit value for benzene ( $5 \mu g/m^3$ ), were limited to very few locations in Europe in 2012, but 10% to 12% of the EU-28 urban population was still exposed to benzene concentrations above the estimated WHO, World Health Organization (2000) reference level ( $1.7 \mu g/m^3$ ), which is associated to an excess lifetime risk (of leukaemia) of 1/100,000. Exceedances of the limit value are often ascribed to close proximity to industrial or traffic sources.

Moreover, accordingly to the European indoor air quality guidelines for benzene (Jantunen et al., 2011), typical values range between 2 and  $15 \,\mu\text{g/m}^3$  with indoor sources contributing up to 40% to the exposure of citizens, reaching more than 75% for most severe exposures (up to  $50 \,\mu\text{g/m}^3$ ).

In this context the recent French Decree (2011) provided a 5  $\mu$ g/m<sup>3</sup> guideline value for benzene for long-term exposure entered into force on 1 January 2013 which has to be lowered to 2  $\mu$ g/m<sup>3</sup> on 1 January 2016. The provided value is related to carcinogenic hematological effects due to life-time exposure to benzene in indoor air coming from indoor sources (smoke, fireplace, furniture, etc.) but also from outdoor air sources (traffic, heating, industry, etc.). The French Decree followed the opinion of the French Agency for Environmental and Occupational Health Safety relative to the proposal of indoor air quality guideline values for benzene (Afsset, French Agency for Environmental and Occupational Health Safety, 2008).

Taking into account the information highlighted above, some concern may emerge for outdoor/indoor relationship in inhabited neighborhood of industries.

Coastal cities hosting multiple potentially impacting activities as chemical plants, steel plants, refineries and crude oil tank farms can undergo emissions of VOCs and benzene among these (Ras-Mallorquí et al., 2007; Valerio et al., 2005; Gioda et al., 2004).

Taranto (SE of Italy) (Bruno et al., 2006; Viviano et al., 2005) and Trieste (NE of Italy) are two examples of such cities in Italy. The latter city was object of several studies for what concerns sediments (Barbieri et al., 1999a, 1999b; Adami et al., 1997) and air particulate matter (Cozzi et al., 2009, 2010, 2012; Astel et al., 2010). In particular polycyclic aromatic hydrocarbons (PAHs) were already found in anomalous concentrations in sediment sampled close to the steel plant (Adami et al., 2000).

Previous studies on BTEX concentrations in the city of Trieste, measured from 2001 to 2008 (Astel et al., 2013) showed that on an overall city scale, vehicular traffic seemed to be the most relevant source of environmental benzene contamination; during the considered timeframe a clear improvement of air quality related to this parameter was shown.

Despite of that, anomalies in the concentration of outdoor benzene were detected in Trieste during 2010 and 2011 by the regional environmental protection agency (ARPA FVG, 2011, 2012) in the framework of a monitoring program including also a station aimed at evaluating fugitive emissions from a steel plant in Trieste and positioned close to the works boundary for fence monitoring. Representetiveness of the concentration measured in that station for exposure of civil population was questioned in those public reports.

Coke oven batteries, which are a part of the steel plant, are a renown source of fugitive emissions of benzene and other volatile organic compounds beside particulate matter and semivolatiles (Shi et al., 2015; EMEP-EEA, 2009; Aries et al., 2007).

Several studies have been produced regarding workers exposition (Chang et al., 2010; Tsai et al., 2008; Bieniek et al., 2004). Only a few number of studies concerned VOCs exposure at or out of the boundary of an integrated steel plant (Ciaparra et al., 2009; Valerio et al., 2005; Thomas, 1990) and they showed high or low values but they were often limited in time.

As WHO, World Health Organization (2000) recommended a wise choice of monitoring stations positioning in proximity of "hot spots" to define and assess the representativeness of each site paying attention to micro-scale conditions, we focused our work on the district of Servola in the city of Trieste (NE Italy) where an integral cycle steel plant is located. A one year monitoring campaign (from spring 2012 to spring 2013) has been carried out to evaluate the concentration of BTEX in outdoor and indoor air in five houses far up to 1100 m from the coke oven batteries.

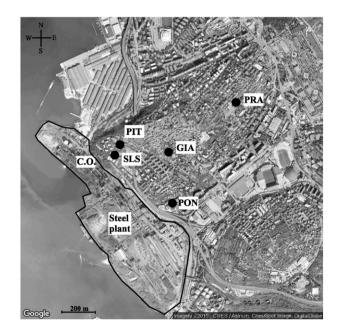
To our knowledge, no such a "small-scale" study focused on both outdoor and indoor air sampled in inhabited buildings near an integrated steel plant has been published in scientific literature to date.

#### 2. Materials and methods

#### 2.1. Sites description

Trieste is a city located in NE Italy by the Adriatic Sea counting about 200,000 inhabitants. In the Servola district (about 12,000 inhabitants – density 8300 inh/km<sup>2</sup>) an integral cycle steel plant is present. The dwellings of the district are positioned in very close proximity of the plant. The steel plant overlooks the sea to the west and the inland to the east.

Five locations positioned at an increasing distance from the plant were selected for BTEX sampling outdoor and indoor of inhabited buildings (Fig. 1). Sampling locations were named by a short three letters identifier: Via S. Lorenzo in Selva is SLS, Via Giorgio Pitacco is PIT, Via dei Giardini GIA, Via del Ponticello PON and Via Marco Praga PRA. Table 1 reports on the distance of the sampling locations from the center



**Fig. 1.** Map of Servola district in Trieste (NE of Italy). The five sampling sites (SLS, PIT, GIA, PON, PRA), the boundary of the steel plant and the position of the coke oven batteries (C.O.) are highlighted.

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