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Identification of stationary sources of air pollutants by concentration statistical analysis

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ABSTRACT

The atmospheric concentrations of benzene, toluene, ethylbenzene and isomeric xylenes (BTEX) in a medium-sized town (S. Maria Capua Vetere, about 32000 inhabitants, Southern Italy) have been determined during working days and weekends in 2006. The procedure used was 24 h passive adsorption by samplers distributed throughout the town followed by GC/MS analysis. On a yearly base, the arithmetic mean benzene concentrations were above the limit required by the 2000/69/CE European Directive. The Pearson correlation coefficients of the 24 h geometric mean BTEX concentrations were indicative of stationary sources of toluene located in a well circumscribed area of the urban territory, active only during the working days and not officially recognized. The results highlight the effectiveness of the statistical approach used in this study for the identification of pollutant sources.

1. Introduction

Aromatic hydrocarbons, in particular benzene, toluene, ethylbenzene and xylenes (BTEX) are among the main pollutants in urban areas (Seinfeld and Pandis, 1998) and their adverse effects on human health are well documented (Victorin, 1993). Several studies (Hinwood et al., 2006; Hinwood et al., 2007; Kerbachi et al., 2006; Martins et al., 2007; Yassaa et al., 2006) have shown that the major contribution to BTEX in urban areas is from the vehicular traffic. Organic compounds emitted from the exhaust pipe consist mainly of unburned aromatic fuel components (Siegl et al., 1992). The air content of benzene relative to other aromatic compounds is significantly higher than its fuel percentage because benzene is a partially oxidized product of other aromatic compounds (McGinty and Dent, 1995). The air concentration of benzene is subject to restriction in many countries whilst for the other BTEX the norms are usually more permissive. The Directive 2000/69/CE of the European Union has fixed the limit benzene concentration to 5 $\mu g\, m^{-3}$ for the urban atmosphere.

Besides vehicular traffic, emission from stationary sources, mainly factories, also plays a relevant role in affecting the atmosphere quality (Sharma et al., 2007; Srivastava, 2004). Volatile organic compounds (VOCs) including BTEX are widely used in industries and their emission in the atmosphere is a constant risk for the population. Fuel stations, small factories, maintenance shops of house-hold appliances and garages are the main urban stationary sources of VOCs. The assessment of vehicular and non-vehicular contribution to VOCs is an indispensable step for the enforcement of measures directed at improving the quality of urban air. Certain VOCs assumed to be emitted exclusively from vehicles have been used as indicators of vehicular pollution, for example methyl *tert*-butyl ether, an octane-enhancing gasoline additive (Chang et al., 2006). This approach, however, should be used with caution, as the relative contents of BTEX compounds in the atmosphere change with the aging of pollutants, because of their different degradation rate (Brocco et al., 1997). Other factors affecting VOCs concentrations include the car speed and notably the diffusion and efficiency of catalytic converters (Heeb et al., 2000).

In the present study vehicular and stationary emissions in a medium-sized town in Southern Italy were assessed by a novel procedure based on the statistical analysis of spatial and temporal variations of BTEX concentrations.

2. Experimental

2.1. Sampling sites

The BTEX concentrations have been determined in the atmosphere of S. Maria Capua Vetere, about 32000 inhabitants, in Southern Italy (lat. 41° 5′ N, long. 14° 15′ E).

The location of the sampling sites is indicated in Fig. 1. The sampling was performed in working days and on weekends throughout 2006, except August. We performed 1 or 2 samplings per month

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Fig. 1. Map of the sampling points in S. Maria Capua Vetere.

in working days and six samplings on weekends for the whole year, from Saturday evening to Sunday evening. The data relative to rainy days were not included. All sampling points were located along the streets of the city, about 5 m above the ground level and not in proximity of cross-roads.

2.2. Analytical procedures

The BTEX compounds (benzene, toluene, ethylbenzene, o-xylene and (m+p)-xylene) were collected by Radiello® diffusive samplers, patented by Foundation Salvatore Maugeri (FSM). The exposition time for each monitoring campaign was 24 h. The samplers consisted of a stainless steel net cylinder with a 100-mesh-grid opening, packed with 530 mg of activated charcoal. The analyses were carried on a Perkin Elmer GC–MS instrument, model Clarus 500, equipped with a thermal desorption instrument, model TurboMatrix ATD. The analytes, extracted from exposed cartridges by a stream of helium (80 ml min^{-1} at 320 °C for 10 min), were condensed in a trap at 2 °C and vaporized by heating at 290 °C for 1 min.

The column used was an Elite-5MS fused silica capillary, 30 m × 0.25 mm, 0.25 μm film thickness (Perkin Elmer Instruments). The oven temperature program was 35 °C for 5 min, to 120 °C at 8 °C min^{-1} and to 200 °C at 15 °C min^{-1}. The carrier gas was He at a flow rate of 1 ml min^{-1}. The mass spectrometer scanned from 35 to 300 a.m.u. every 0.1 s, in the electronic impact (70 eV) mode. The ion source temperature was 180 °C and the multiplier voltage 350 V.

2.3. Statistical analysis

Statistical analysis was carried out using the statistical software package Statistica 7 of StatSoft.

3. Results and discussion

3.1. Concentrations in the sampling sites

Although this survey was carried out in a medium-sized town, the geometric mean concentrations were relatively high with