

THE LIFE+ POPULATION EXPOSURE TO PAH (EXPAH) PROJECT: INDOOR/OUTDOOR MONITORING AND EMISSIONS ESTIMATION IN ROME

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ABSTRACT

Polycyclic Aromatic Hydrocarbons (PAHs) are a class of pollutants of increasing concern for their occurrence in the environment. There is evidence that PAH exposure is related to lung, skin, and bladder cancer in humans. In highly urbanized areas domestic heating and mobile sources are the largest contributors of PAHs. The EC funded a project (EXPAH "Population Exposure to PAHs") under the LIFE+ program aims to quantify the population exposure to PAHs in urbanized areas, and to determine the impact on human health. The project integrates measurements, modelling and epidemiologic methods to achieve the above goals. The city of Rome was chosen as study area. The current paper presents the aims, the methodology and preliminary results of the ongoing study.

1. INTRODUCTION

Polycyclic Aromatic Hydrocarbons (PAHs) are a class of complex organic chemicals of increasing concern for their occurrence in the environment. They are ubiquitous in ambient air and some of them have been identified as suspected carcinogens (IARC, 1983). PAHs can be found in the atmosphere in both gaseous and particulate forms (PM_{2.5} and PM₁₀). The best known PAH is the benzo[a]pyrene (B[a]P). PAHs and their derivatives are produced by the incomplete combustion of organic material. In general there are five major emission sources of PAHs: domestic, mobile, industrial, agricultural and natural. In highly urbanized areas domestic heating and mobile sources, and specifically vehicles, are the largest contributors of PAHs (Zhang and Thao, 2009). There is evidence that PAH exposure is related to lung, skin, and bladder cancer in humans. DNA damage induced by PAHs exposure was demonstrated by different authors (eg. Garte et al., 2007). Long-term exposure to PAHs has been associated with gene mutation cell damaging and increased risks of cardiopulmonary mortality. Short term exposure to PAHs has been reported to cause impaired lung function in asthmatic people and thrombotic effect in people affected by coronary heart disease. Given the outlines illustrated above, the LIFE+ EXPAH project will improve the knowledge concerning population exposure to PAHs and their related health effects.

2. THE POPULATION EXPOSURE TO PAH EXPAH PROJECT

2.1 Aim and objectives

In order to address the environmental and health problems induced by emission, dispersion and transformation of polycyclic aromatic hydrocarbons (PAHs) compounds, EC funded the "Population Exposure to PAH" (EXPAH) project (Oct 2010-Dec 2013; <http://www.ispe.it/expah>) under the LIFE+ programme. The overall goal of the project is to identify sources and quantify population exposure of special segments of population, namely children and elderly people, to particulate PAHs in highly urbanized areas and to assess the impact on human health. An integrated approach, based on measurements and modeling techniques, will be used to estimate the spatial distribution of population exposure to PAHs, to identify key determinants of high exposures including time-activity and locations in relation to the sources and to estimate potential health effects on the target population.

More specifically the EXPAH objectives are:

- to develop a state-of-the-art PAH emission inventories for the test city;
- to improve and integrate air pollution models to describe the emissions, diffusion, atmospheric transformations, and removal of transport-related pollutants with particular relevance to PAHs;
- to estimate the actual concentrations of particulate matter and PAHs in different areas of the city, using existing monitoring network and field studies, to assess the human exposure in different living places;
- to develop an outdoor-indoor infiltration model able to estimate the amount of both particles and PAHs of ambient origin present in each predefined indoor microenvironment;
- to estimate the mean exposure of the target populations to particles and PAHs using data derived from the air pollution model, infiltration models and population time-activity patterns through an exposure model;

- to estimate the short-term and long-term health impact of particles and PAHs on mortality and morbidity using actual and model estimated exposures levels;
- to improve our understanding of the human health effects and the corresponding exposure-response relationships of PAHs in order to evaluate the impact of different threshold values of air pollutants concentration, with special attention on organic compounds;
- to evaluate the health impact in view of existing, planned as well as alternative future EU policies in order to provide recommendations for adaption and mitigation strategies.

The study area is the city of Rome experiencing serious pollution problems, particularly for PM₁₀, NO₂ and O₃. Several studies have reported health effects due to air pollution in this city (e.g., Forastiere et al. 2008) and an increase of 10 µg/m³ of PM₁₀ is expected to be responsible for an increase of 1.1% in cardiovascular and respiratory mortality. Moreover, exposure and health effects to PAHs have not been investigated.

2.2 Methodology

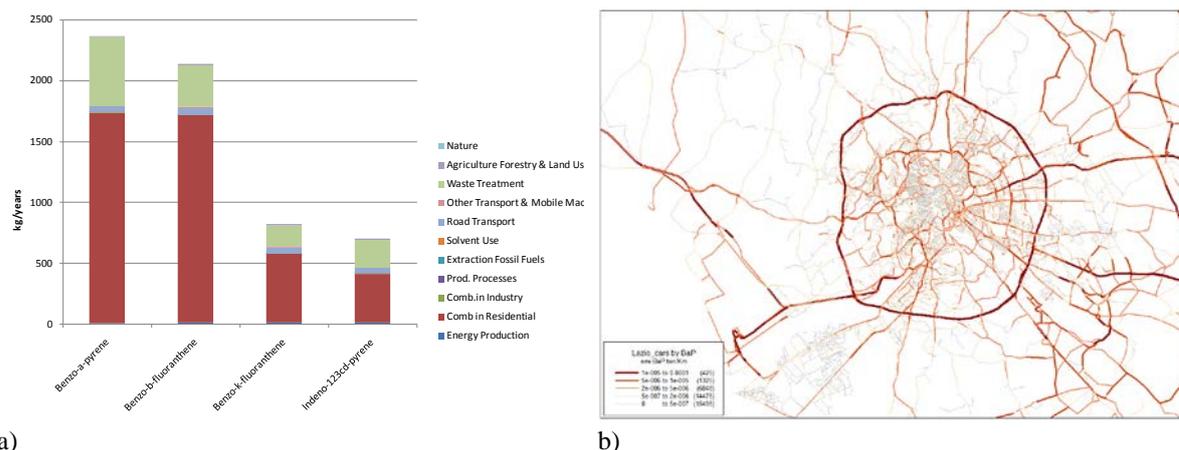
The EXPAH project implements 27 actions. An emission inventory has been developed starting from the available national and local inventories. It will be provided to the Chemical Transport Model (CTM) based on the meteorological model RAMS and on the FARM model (Silibello et al., 2005), to estimate the ambient particles concentration and their PAHs content. Principal gaseous reactions of PAHs, as well as their partitioning between gaseous and particulate phases species will be included. Surface and upper air meteorological measurements are on going to provide data to RAMS model for assimilation and validation.

Field campaigns for characterization of the PAHs particle are in progress as a supplement to ongoing local air quality monitoring. Seasonal indoor-outdoor measurements are on going at different living places (e.g. home, office, school, car; hereafter microenvironments) to evaluate the ability of ambient PAHs to penetrate and pertain to these microenvironments. These results will be then used to develop an infiltration model. Personal exposure monitoring is carrying out for people living in selected environments to evaluate model results and to quantify actual PAHs exposure. In order to estimate the exposure of the target population, a model based on a methodology developed under the EU FP5 FUMAPEX project (<http://fumapex.dmi.dk>, Baklanov et al., 2007) will be used. Maps of population exposure will be set up, starting from PAHs concentrations, as provided by the CTM model, by applying a weighted average concentration on the basis of both the amount of time spent in each microenvironment and of the estimated concentration in each microenvironments, as provided by the infiltration model. Time activity data for the target population will be collected to support the exposure model. Seasonal and annual PAHs and PM_{2.5} exposure cumulative distribution will be calculated. Health effects will be then estimated in terms of short and long-term increases in mortality and morbidity related to air pollution exposure in the Rome population, using the results of the air pollution and exposure models. In particular, an evaluation of the impact on mortality considering the specific time period, the area of the city and the source will be conducted. The impact on hospitalization as well as the impact of long term exposure on the occurrence of lung cancer and other cancer forms, coronary and cerebrovascular events will be evaluated. Finally, an attempt to improve our understanding of the human health effects and the corresponding exposure-response relationships of PAHs will be carried out. A number of possible emission scenarios will be then evaluated to investigate the possible health impact attributable to planned as well as alternative future EU policies and international commitments.

3. EXPAH RESULTS

3.1 PAHs emission estimation in Rome

A reference PAHs emission data set has been constructed on the basis of emission inventories available at national and international level. Due to the specific objectives of EXPAH project, it has given preference to emissions with high space resolution over the target area of Rome. This aim has been fulfilled starting from the national emission inventory ISPRA2005 characterized by province level resolution and its downscaling at municipal level resolution INCOM2005. Those inventories include total PAH emissions for each sources sector but they do not include information on the different congeners. Emissions of the 4 PAHs identified in the UNECE POPs protocol (benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene and indeno[123-cd]pyrene) have been estimated by means of the profiles available in literature for the various emission sectors. The results have been analyzed and compared with the European scale emission inventory developed and provided by TNO. The inter-comparison highlights the large degree of uncertainty that affects PAHs emissions and that can generally be considered larger than that associated to other pollutants. Details on the PAHs emission inventory and inter-comparison analysis can be found in a specific technical report available at project web site. As an example, the figure 1 shows the speciated contribution of the different source sectors to PAHs emission inventory for the Lazio region. The analysis of the emissions inventory of Lazio region and Rome metropolitan area confirmed that combustion in residential heating is the main source of PAHs accounting for 73% of emissions in Lazio region, growing to 92% within Rome municipality.



a) Figure 1. (a) Speciated contribution of the different source sectors to PAH yearly emissions in Lazio region in the year 2011 for different sources; (b) Benzo(a)pyrene yearly emissions from cars as estimated from traffic flow in the Rome road network.

Waste treatment contribution is the second main contribution with 22% of emissions over the Region, 9% over the Province and 3% over Rome municipality. Road transport contribution remains in the 3-4% range over the different considered areas. Detailed traffic PAHs emissions have been calculated for city of Rome and its Province. Vehicles flow for each link of Rome road network has been estimated by the local mobility authority, by means of a traffic assignment model representing the traffic flows on the road network, on the basis of origin–destination (OD) matrices and observed traffic data. Hourly emissions of all the pollutants, including PAHs, have been calculated for each road link and each vehicles class by means of TREFIC model (Nanni et al., 2005), based on the COPERT IV methodology. Figure 1b shows the yearly emissions of Benzo[a]pyrene for cars. As expected the largest part of emissions are produced in the city of Rome.

3.2 PAHs exposure in the Rome metro lines

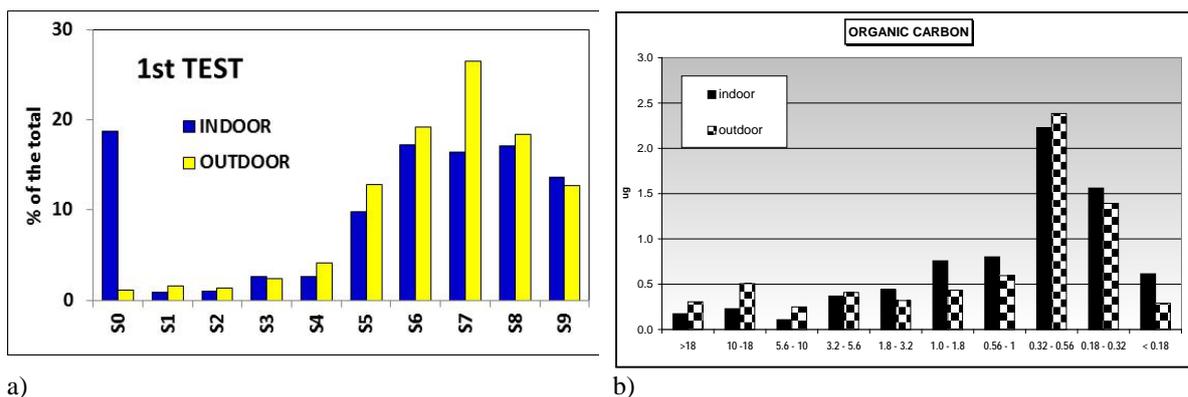
In order to evaluate the PAH exposure of population in different microenvironments, personal exposure measurements were carried out in the metro lines (A and B) of Rome and in the shopping area of Termini railway station. Samplings were carried out during three different days for 8 hours, by using a pair of SKC low volume sampler, each one equipped with a PM_{2.5} inlet (breathable fraction). Sampled particles were processed according to the procedure based on solvent extraction, clean-up through column chromatography and CGC-MSD analysis. Table 1 shows the measured PAHs exposure. Detailed results can be found in a technical report available at project web site.

Table 1. PAHs personal exposure concentrations (ng/m³) in Metro of Rome and in Termini shopping center

Compound	Metro April 29th, 2011	Termini shopping centre May 11th, 2011	Metro May 12th, 2011
Sampled volume (m ³)	1.47	1.73	2.06
benz(a)anthracene	0.46	0.18	0.41
chrysene+triphenylene	1.05	0.46	1.13
benzo(b/j)fluoranthene	0.88	0.34	0.95
benzo(k)fluoranthene	0.32	0.16	0.32
benz(e)pyrene	0.39	0.23	0.44
benzo(a)pyrene	0.11	0.11	0.15
indeno(1,2,3-cd)pyrene	0.23	0.16	0.18
benzo(ghi)perylene	0.28	0.25	0.22
dibenz(a,h)anthracene	0.09	0.04	0.04
<i>total PAHs</i>	<i>8.9</i>	<i>2.6</i>	<i>4.5</i>

3.3 PAH congener partition among the size fractions of aerosols.

Two experiments have been carried out in the CNR-IIA buildings. Particulates were collected by means of a MOUDI-100 cascade impactor (MSP Corporation, MN, USA) running at 30 L/min, equipped with nine cut-off lines and one end-point filter collecting residual particles. The 50%-efficiency cut-offs were fixed at 18 (inlet), 10, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18 μm. Samplings were performed in parallel indoor (inside an office room) and outdoor and lasted seven consecutive days. The experiments were made from April 26th to May 3rd, 2011 and from May 3rd to May 10th, 2011. The PM fractions (labelled from S0 up to S9, in the order) were collected on pre-baked 47-mm o.d. quartz membranes for PAHs, elemental and organic carbon analysis.



a) Figure 2. Percent distribution of total PAHs (a) and mass of organic carbon (b) in the size-segregated suspended particulate fractions. Montelibretti, April 26th – May 3rd, 2011(a). Symbols: S0 >18 μm; 10 μm ≤ S1 ≤ 18 μm; 5.6 μm ≤ S2 ≤ 10 μm; 3.2 μm ≤ S3 ≤ 5.6 μm; 1.8 μm ≤ S4 ≤ 3.2 μm; 1.0 μm ≤ S5 ≤ 1.8 μm; 0.56 μm ≤ S6 ≤ 1.0 μm; 0.32 μm ≤ S7 ≤ 0.56 μm; 0.18 μm ≤ S8 ≤ 0.32 μm; S9 ≤ 0.18 μm.

The distribution of all PAH congeners is similar indoors and outdoors, unlike the indoor environment is “sealed” for long time and prevented from outer contamination. In particular, BaP accounts for ~4.2-5.5% of total PAHs at both sides. EC/OC results show that the dimensional distribution of these species is in the finest range, with maximum in the interval 0.32 – 0.56 μm for organic carbon and bimodal distribution with maxima in the ranges below 0.18 μm and between 0.32 and 0.56 μm for elemental carbon. Outdoor-indoor differences higher than 1 are observed only for EC in the size range between 0.32 and 1.8 μm. Detailed results can be found in a technical report available at project web site.

4. CONCLUSIONS

The aim, objectives and methodology of the EU LIFE+ EXPAH project has been presented. After the first of three years of project life, it provided the first results about PAHs emission inventory in the city of Rome as well as PAHs personal exposure in the metro and data concerning both indoor and outdoor PAHs and EC/OC distribution in the size-segregated particulate fraction. Combustion in residential heating is the main source of PAH accounting for 92% of emissions within Rome municipality. An exposure of 4-9 ng/m³ of total PAHs was found in the metro, while similar size distribution of PAHs was found in indoor and outdoor with a peak located in 0.32-0.56 μm fraction.

5. REFERENCE

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