PAHs environmental and health effects analysis, methodology employed, and its level of portability in other EU areas

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Summary

Introduction

The present report deals with the brief description of the methodologies employed in the EXPAH project to estimate PAHs population exposure, health effects, and the level of portability of the methods used. After a short introduction on the environmental target problem, an overview of the design of the EXPAH project and its methodologies are presented. Then the level of portability of employed methodologies and data used are analyzed and discussed. Conclusions and remarks are finally presented.

1. PAHs environmental problem

Among Persistent Organic Pollutants (POPs), Polycyclic Aromatic Hydrocarbons (PAHs) are a class of complex organic chemicals of increasing concern for their levels in the environment. They are ubiquitous in the atmosphere where are present as gases or enfolded in particulate matter ($PM_{2.5}$ and PM_{10}) on the basis of their volatility and chemical structure. PAHs and their derivatives are produced by the incomplete combustion of organic material: mainly from anthropogenic combustion and partially from biomass burning. In general there are five major emission sources of PAHs: domestic, mobile, industrial (including power generation and waste processing), agricultural and natural. In highly urbanized areas domestic heating and mobile sources, specifically vehicles, are the largest contributors of PAHs, with diesel fuelled cars releasing higher particulate emissions than gasoline fuelled ones. As a consequence populations living in urban areas are exposed to pollutants which have potential health effects.

The United States Agency for Toxic Substances and Disease Registry consider 17 priority PAHs on the basis on their toxicological profile. The selection criteria were : availability of information, suspicious of harmful effects, greater exposure of population, higher ambient concentrations than other PAHs. The best known PAH is the benzo[a]pyrene (B[a]P) which has been classified as carcinogen for humans (group 1) by IARC. Some other PAHs have been identified as probable (benz[α]anthracene and Dibenz[a,h]anthracene) or possible (benzo[bjk]fluoranthene, benzo[c]phenanthrene and indeno[1,2,3-cd]pyrene) carcinogens.There is strong evidence for the relationship between PAH exposure and lung, skin, and bladder cancer in humans. DNA damage induced by PAHs exposure was demonstrated by different authors and a genotoxic risk was reported in people working in urban ambient. Long-term exposure to PAHs has been associated with an increased risk of cardiopulmonary mortality . Short term exposure has been associated with impaired lung function in asthmatic people and thrombotic effect in people affected by coronary heart disease.

The European Directive 2004/107/EC proposed a target value of 1 ng/m^3 B[a]P for the total content in the PM10 fraction averaged over a calendar year. This directive recommended also to assess the specific contribution of B[a]P and to monitor other relevant PAHs, with the aim of supporting policy actions. However, PAHs are not always continuously monitored in each Member State. Consequently the European population exposure to PAHs and their time and spatial characteristics are not well known.

The LIFE+ EXPAH project has filled this gap of knowledge producing useful information to assess the PAHs exposure of urban population and its related health effects.

2. Overview of EXPAH project

The EXPAH project addressed the environmental and health problems induced by emission, dispersion and transformation of PAHs compounds. The overall goal of the project was to identify and to quantify population exposure to PAHs content in particulate matter and to assess the impact on human health, in the urban area of Rome, in order to support environmental policy and regulation. An integrated approach has been used to estimate the spatial distribution of PAHs, both measurements and modeling, to identify key determinants of exposures including time-activity, indoor-outdoor exposure and living near high traffic roads, and to estimate health effects. EXPAH project built a prototype method to provide an integrated knowledge on PAHs from concentrations to health impact in urban areas and to define scenarios achievable by hypothetical reductions of exposure. The project can contribute to implement and evaluate environmental policies at both local and national levels. The results can also provide a support to EC legislation concerning PAHs.

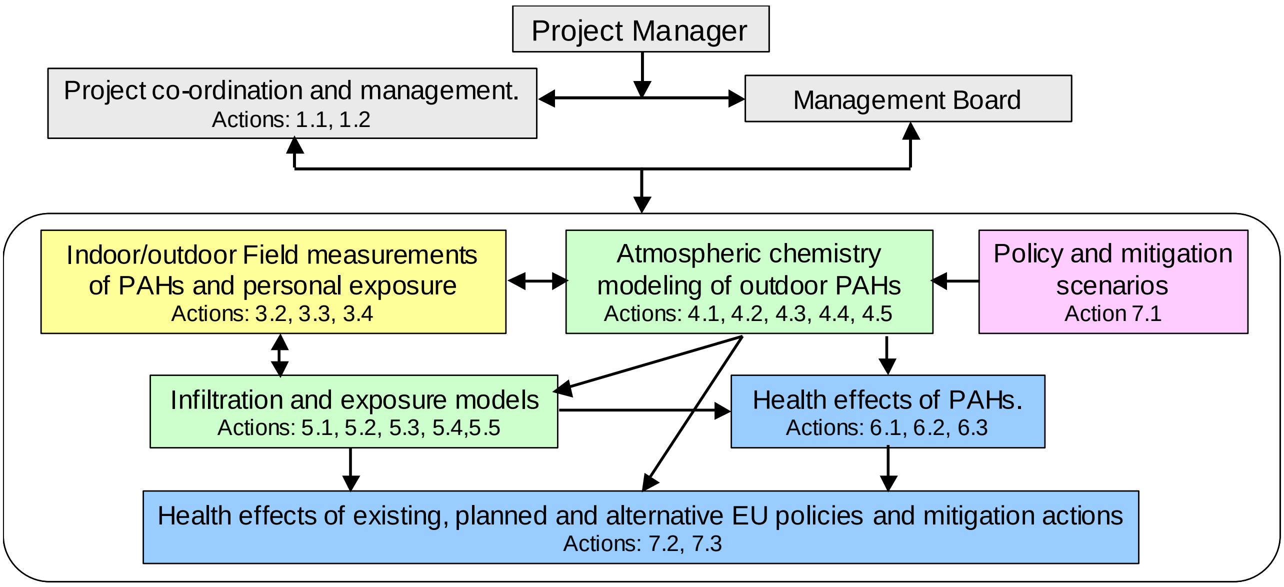


Figure 1Layout and interconnection of EXPAH actions

3. The EXPAH methodology

The EXPAH methodology consists in an integrated approach which involves modeling PAHs ambient concentrations, starting from emission inventory and properly validated by dedicated measurements, and assessing epidemiologic estimates of short-term and long-term health effects of PAHs. (fig. 2)

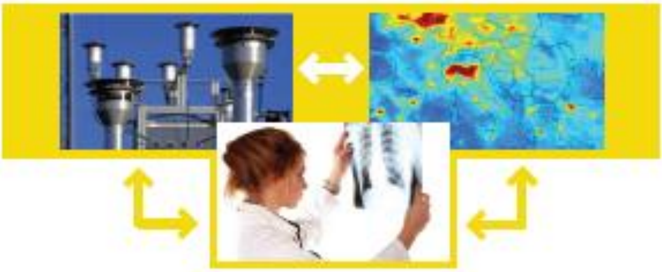


Figure 2 The integrated approach which involves measurements, modeling and epidemiologic investigations.

The potential effects on air quality induced by potential EU and local policies are then evaluated by means of the modeling chain simulating emission, dispersion and transformation processes and using a “What-If” scheme where changes in emissions are evaluated in terms of effects produced. Figures 3 and 4 show the diagrams of the exposure assessment methodology and the exposure-health chain.

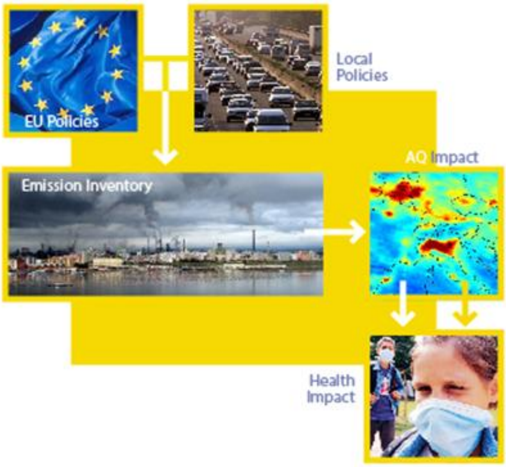


Figure 3. Diagram of the exposure assessment methodology.

In the EXPAH project the health effects are evaluated by considering all involved processes. Starting from the emissions of the main identified sources, the ambient PAHs concentrations are estimated by means of modeling techniques, properly validated by dedicated measurements.

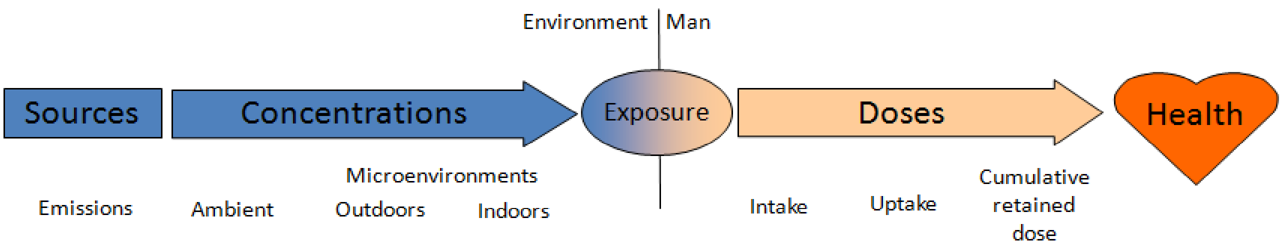


Figure 4. Exposure-health chain.

As population is known to spend most time in indoor environments, a proper assessment of exposure has to consider the indoor infiltration of outdoor (ambient) pollutants. The EXPAH project availed also of specific methods to address these aspects of exposure: a model to estimate indoor/outdoor infiltration and indoor concentrations in the main living environments, and a sample survey to assess microenvironment daily exposure to PAHs. The overall exposure has been estimated by summing all single exposures experienced in the visited living environments weighted for the time spent in each of them.

Figure 5 shows the modeling methodology to assess the PAHs exposure. Whenever this methodology is applied, starting from modeled environmental PAHs impact, urban maps of population exposure to PAHs can be obtained and then used to evaluate health effects.

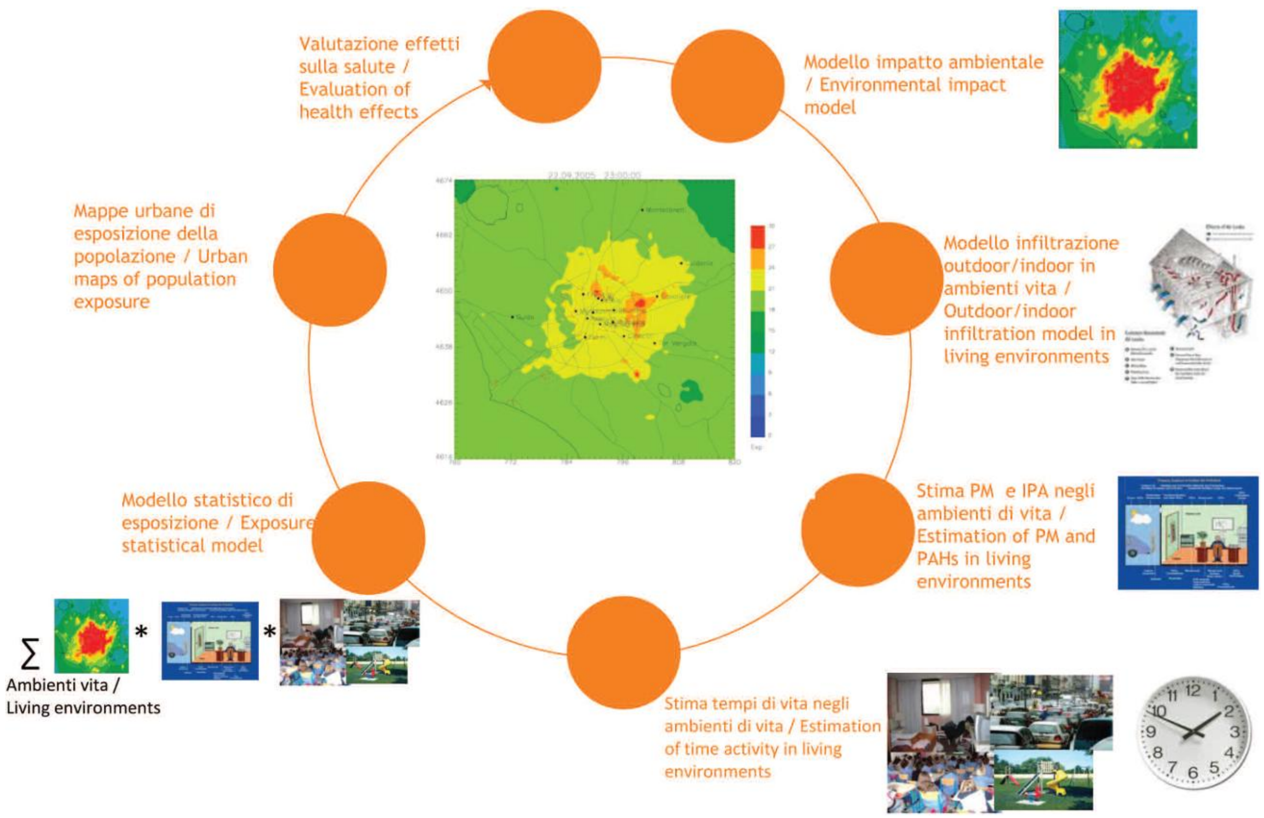


Figure 5. Modeling methodology to assess PAHs exposure.

The methodology has been applied to the city of Rome, which is one of the largest cities in Italy covering an area of 1290 km^2 with a population of about 3 million inhabitants.

A brief description of the methods to address specific exposure characteristics follows .

3.1. Estimates of time activities and microenvironments exposures

The EXPAH project aimed to collect and analyze time activity data of population groups to get information about which environment (home, school, car bus, outdoor, etc) are attended during weekdays and week-ends . The PAHs exposure experienced by population during its typical day, were evaluated, in Rome as a weighted exposure of the different pollutants levels experienced in the different micro-environments. Both children and elderly people (700 children (7- 8 years old) (GASPII study) and 998 elderly (65-85 years old) (IMCA study)) were interviewed by questionnaire to collect time activity data on a seasonal basis and for two days of the week, Sunday (public holiday) and Wednesday (week-day).

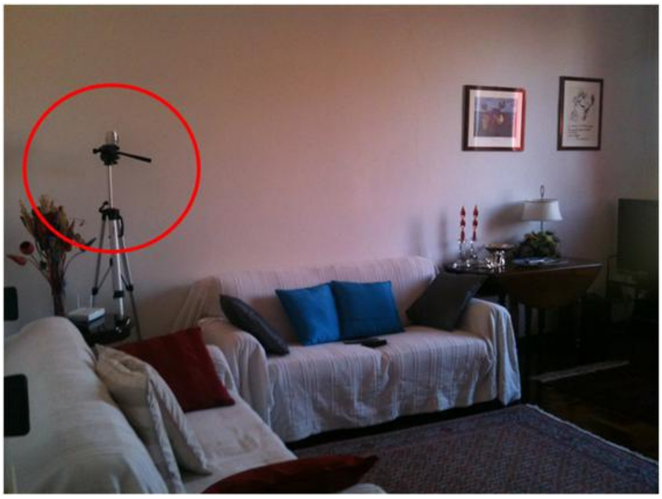
Two kind of analysis of the data were performed: a time slot analysis (by half-hour periods) and a daily analysis. For the time slot analysis we calculated the half an hour distribution of the time activities in percentage, stratifying by season (spring/summer and autumn/winter) and day of the week (public holiday and week-day), in the total population and in males and females, separately. For the daily analysis, we calculated the daily distribution of the time activities in percentage, stratifying by season (spring/summer and autumn/winter), day of the week (public holiday and week-day) and gender (males and females).

The most visited micro-environments were identified as well as their daily occupation rates for children and elderly people in two seasonal periods and day of week. Details about the methodology and results can be found in an EXPAH technical report (Porta, 2012).

3.2. PAHs monitoring of indoor and personal exposure

Several air pollutants (PAHs, $PM_{2.5}$ with chemical composition, gases) have been measured in different living/working microenvironments, in Rome, in the period 2011-2012 to support modeling studies on pollutants infiltration factors. The methodology was based on low volume active sampling on PTFE filters and gas chromatography/mass spectrometry determination of $PM_{2.5}$-bound PAHs non-volatile congeners. Filters were aggregated on weekly bases. According to the study design, two in-field campaigns in summer and in winter-spring were performed in 20 living/working environments including two cars, one bus, six schools, two offices and nine houses, both indoor and outdoor. In addition, data on concentrations of $PM_{2.5}$ and associated PAHs were monitored by three urban fixed stations (Air Pollution Regional stations) during the in-field campaigns.

(a)



(b)

Figure 6. Examples of in-field sampling at schools (a) and houses (b).

The PAHs analyses focused on the carcinogenic ones, namely benz(a)anthracene, benzo(b)fluoranthene, benzo(j)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, dibenz(a,h)anthracene and the mutagenic benzo(ghi)perilene.

PAHs concentrations were higher in winter respect summer. The indoor PAHs concentrations were not negligible, but the influence of indoor sources was scarce in the monitored environments. A good correlation indoor/outdoor of the PAHs concentrations was found, PAHs infiltration factors were derived for the monitored living environments by applying a best linear fit between outdoor and indoor concentrations. Table 1 shows the obtained infiltration factors for the monitored microenvironments.

Table 1. PAHs and B[a]P infiltration factors for the monitored microenvironments.

	Infiltration factor	
Micro-environment	**PAHs**	**B[a]P**
Home	0.66	0.69
School	0.67	0.63
Office	0.56	0.69
Car	0.59	0.61
Bus	0.92	0.93

Personal exposure has been estimated in five children and four elderly A 24-hour sampling activity has been performed during two 15 days campaigns by means of personal samplers, battery operated, positioned to sample the inhaled $PM_{2.5}$. The samplers operated at flow-rate of 10 L/min and using Teflon filters. The exposure was found to mainly occur in indoor environments, such as schools and houses, where people stayed longer; no significant differences were observed between elderly and children.

Details on methodologies and PAHs, PM2.5 results can be found in found in Romagnoli et al. (2014), Gatto et al. (2013) and in the EXPAH technical report (Cecinato et al., 2013).

3.3. PAHs modeling and assessment of environmental and population exposure impacts

The three dimensional Eulerian chemical transport model FARM, which accounts for the transport, chemical transformation and deposition of atmospheric pollutants has been used to reconstruct PAHs concentrations, in Rome, during 2011-2012. FARM has been applied to two nested domains: a larger one including Lazio Region (61 x 51 grid cells with an horizontal resolution of 4 Km) and a target one including Rome metropolitan area (60 x 60 grid cells with an horizontal resolution of 1 Km). The simulated period lasted from June 2011 to May 2012 (one year). Emission data, which are the main input for FARM model, derived from the Lazio Region inventory and the national emission inventory ISPRA2005 (base-case scenario). PAHs emissions were temporally, spatially and chemically disaggregated. Meteorological fields were provided to model by a prognostic-diagnostic meteorological models system (RAMS-SURFPRO) driven by larger scale meteorological analysis (ECMWF) and local data collected during a proper field campaign. Figure 7 shows a diagram of the modeling system used in EXPAH.

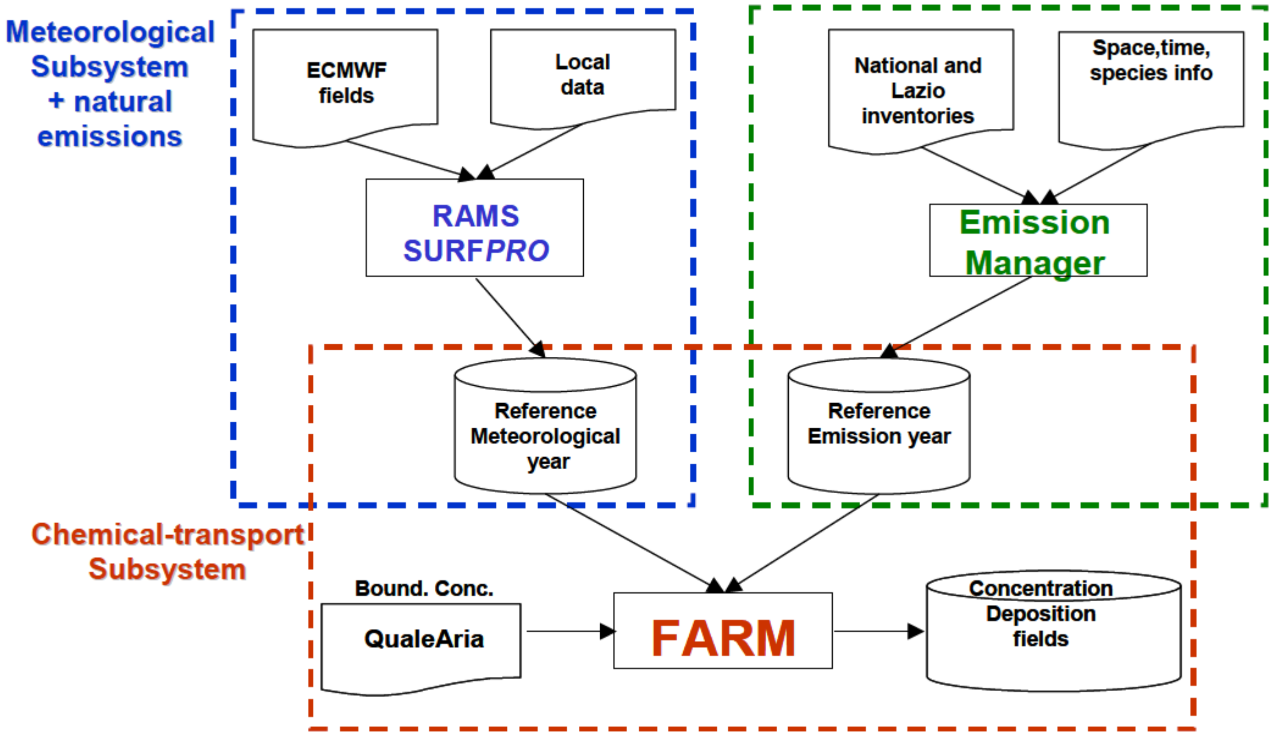


Figure 7 Diagram of the modeling system.

The indoor concentrations were calculated by applying the infiltration factors described in table 1 while the time-activities assessment of population gave the weighted average concentrations in each micro-environment

Thus, an exposure model has been developed to assess exposure of population living in Rome. using outdoor concentrations provided by a Chemical Transport Model combined with the infiltration factor and the time spent in each micro-environment, to estimate the indoor concentrations and the time-weighted estimates of exposure in each micro-environment. Daily exposures were then calculated for PAHs (as sum of Benzo[a]Pyrene(B[a]P), Benzo[b]fluoranthene (B[b]f), Benzo[k]floranthene (B[k]f) and IndenoPyrene (IP)), Benzo[a]Pyrene) and $PM_{2.5}$ for children and elderly population during the period June 2011 - May 2012. Maps of PAHs, BaP and $PM_{2.5}$ exposure were calculated on daily bases during a whole year for the target population living in Rome. Annual and seasonal averaged results were also calculated. Exposure results were validated against PAHs and $PM_{2.5}$ personal exposure measurements carried out on a few subjects owning to the target population.

Details on models methodologies and results can be found in many EXPAH technical reports (EXPAH, 2012; Gariazzo et al (2014a); Gariazzo et al (2014b); Radice et al. (2012); Silibello et al. (2013)).

3.4. Assessment of health effects of PAHs exposure

There is convincing evidence from scientific literature on the effects of particulate matter, especially fine particles (PM2.5), on mortality for cardiovascular and respiratory causes. Among the most carcinogenic compounds of particulate matter, polycyclic aromatic hydrocarbons, specifically benzo[a]pyrene, have been identified and investigated. For this reason, analysis on the association between short-term and long term exposure to PM2.5, total PAHs, and BaP, and cause-specific mortality or hospital admissions have been developed.

In the frame of the analysis of the short-term mortality effects, all subjects deceased in Rome between June 2011 and May 2102 and residing in the city at the moment of death have been selected. Information on age, gender, season of death, place of death, and underlying cause were retrieved (coded according to the International Classification of Diseases revision 9, ICD-9). Four main groups of death causes were considered: natural (ICD-9: 1-799), cardiac (ICD-9: 390-429), cerebrovascular (ICD-9: 430-438), and respiratory causes (ICD-9: 460-519).

Concerning the analysis of hospitalizations, subjects hospitalized in Rome between June 2011 and May 2102 and residing in the city at the moment of admission were selected. Only ordinary, acute and non-scheduled admissions were considered. For each hospitalized subject, we retrieved information on age, gender, season of hospitalization, and primary diagnosis at discharge (coded according to the International Classification of Diseases revision 9, ICD-9). Seven main groups of diagnoses were considered: cardiovascular diseases (ICD-9: 390-459), ischemic heart diseases (ICD-9: 410-414), heart failure (ICD-9: 428), cerebrovascular diseases (ICD-9: 430-438), respiratory diseases (ICD-9: 460-519), chronic obstructive pulmonary disease (ICD-9: 490-492, 494, 496), and asthma (ICD-9: 493).

Individual exposure to PAHs, BaP and PM2.5 was assigned at residential address by means of the FARM Chemical Transport Model system described above. Additional time-varying variables were collected for the epidemiological analysis which include: air temperature, holidays, summer population decrease, and influenza epidemics.

The short-term effects analyses were carried out using a case-crossover statistical approach with "time-stratified" design for the selection of control days. The statistical analysis was performed with multivariate conditional logistic regression models, also adjusting for air temperature, holidays, summer population decrease, and influenza epidemics. Once the adjustment model was defined, the pollutant was added to the regression model. The lag structure of the association between PM2.5/PAH/BaP concentrations and cause-specific mortality as well as disease-specific hospitalization, were inspected by choosing three cumulative lag structures a priori defined to represent immediate, delayed or prolonged effects: lags 0-1, lags 2-5 and lags 0-5, respectively. Finally, for each exposure/outcome combination, one of these three alternatives was chosen as the "reference lag" for the following analyses.

As for long term analysis, the Rome Longitudinal Study (RoLS), which uses the 2001 Census fixed cohort of Rome (Cesaroni et al. 2013) was used for evaluation of health outcomes. It enrolls all 30+ year old inhabitants of Rome on the census reference day (21 October 2001, about 2.5 million inhabitants) not living in institutions (prisons, hospitals or nursing homes), who were resident in Rome since at least five years. Residents in Rome on the 1 January 2008, who were aged 40 years or more were selected. A follow-up during the period October 2001-December 2012 was conducted by determining the vital statusand retrieving information on deceased individuals. The underlying cause of death (coded according to the International Classification of Diseases revision 9, ICD-9) for deceased subjects was collected from the regional health information system. Residential exposure to PAHs and PM2.5 was estimated, as for short term effects, by means of the FARM Chemical transport model system described above, assuming no change of residence or UFP levels at residence occurred since 1997. Health outcomes were identified non-accidental mortality, mortality for cardiovascular disease (ICD-9:390-459), and respiratory disease (ICD-9:460-519), and incident cases of acute coronary events, stroke, and lung cancer. A Cox regression was used to evaluate the association between pollutants and non-accidental mortality, cardiovascular mortality, and incidence of lung cancer, adjusting for several individual and contextual factors.

All the results have been expressed as percent increases of health outcome, and 95% CI, relative to increments in each exposure equal to its interquartile range.

The evidence of an association of all the exposures with short term natural and respiratory mortality was found, especially at delayed and prolonged latencies, whereas the effects on cardiovascular mortality were somewhat weaker.

Little evidence of an association of any of the exposures with cardio-respiratory emergency hospitalizations was found. There was a weak suggestion of immediate association of PM2.5 with cardiovascular morbidity, and prolonged effect on respiratory morbidity.

Evidence of an association of all the long term exposures with non-accidental and cardiovascular mortality, and incidence of lung cancer and stroke was found.

Details on methodologies and results can be found in EXPAH technical reports (Cesaroni et al., 2014).

3.5. Scenario analysis

The EXPAH project aims at investigating the impact of future emission scenarios on environment and health. To accomplish this aim, the expected PAHs and $PM_{2.5}$ emissions, have to be estimated assuming as realized both the implementation of AQ legislations and the related mitigation actions,. The 2020 Current Legislation scenario was considered (2020 CLE) using the results of the GAINS-Italy project which is able to provide the expected emissions as "base" of future exposure scenarios; it allows also to add regional and local mitigation strategies regarding energy. For pollutants not explicitly considered by GAINS (eg. PAHs) it has been chosen to adopt pollutant emission trends, In particular, PAHs have been projected using PM trends for each single emission sector considered.

Moreover, since CLE scenario foresees a growth of PAHs emissions, while measurements campaigns and preliminary simulations indicated the need to reduce PAH winter-time concentrations in Rome, , the substitution of biomass with natural gas for house heating, was considered as the most important local mitigation measure (2020 CLE Scenario with additional measures (AME)).

Details on the future scenarios can be found in the EXPAH technical report (Radice and Finardi, 2014).

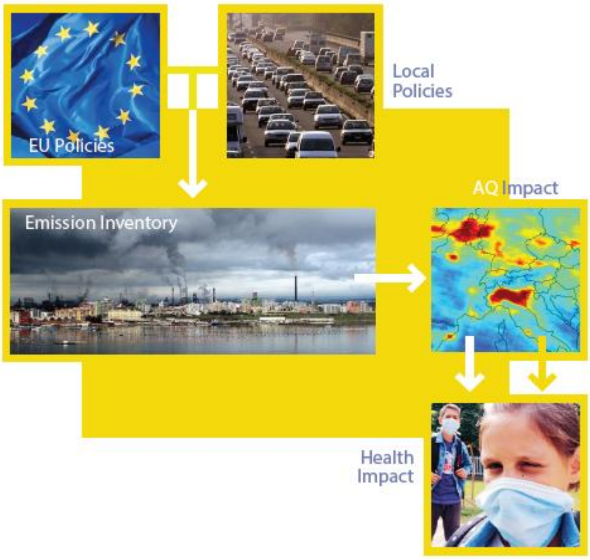


Figure 8. Sketch of exposure assessment in scenarios analysis

Whenever the new 2020 emission scenarios were available at regional and Provincial levels, the methodology already used in EXPAH for the base-case scenario to spatially, temporally and chemically disaggregate the emissions (see par. 3.3) has been reapplied to produce a 2020 scenario over the Rome domain. Then the model FARM has been run again during one year (1^{st} June 2020to 31^{th} May 2020) to obtain the new environmental impacts,.

The impacts of new scenarios on the population exposure have been then estimated by using the 2020 expected levels . while time activities data and infiltration factors were the same used in the base case simulation.

As for health effects, the methodology described above (see par. 3.4), for the long term exposure, was reapplied by using the 2020 expected exposure while keeping health data fixed.

Details on the PAHs environmental impact, population exposure and health effects in the indicated scenarios can be found in the EXPAH technical reports (Silibello et al., 2014; Cesaroni et al., 2014).

3.6. Further possible improvements

Although the EXPAH project adopted the most updated methodologies for the evaluation of PAHs exposure in the urban population, a number of possible improvements in both data and methodologies were identified during this experience.

As for monitoring of PAHs exposure the following improvements can be adopted:

- Better coverage of PAHs temporal profiles by means of more time resolved monitoring or optimized sampling approaches;
- Better technical chemical analysis to increase accuracy and the number of identified PAHs compounds for toxicity and source fingerprint studies;
- More studies on PM2.5 composition and indoor infiltration.
- Assessment of PAHs exposure in living environments with specific indoor sources (kitchen; fireplace; candles; smokers);
- Measurements of micro-climate(eg. temperature, humidity, ventilation, pressure) parameters to better address indoor contaminants behavior;
- Detailed particle size distributions including PAHs related to particle infiltration and respiratory tract deposition;
- Seasonal and source specific variation of PAH compounds according to particle size distribution;

As for modeling techniques the following improvements might be required:

- Better assessment of PAHs emissions with particular attention to biomass combustion sources used as domestic heating;
- Better information on spatial and temporal characteristics of biomass combustion and domestic heating sources;
- Extension of the number of PAHs compounds modeled including their atmospheric transformation processes;
- Extension of PAHs degradation processes in addition to the ozone driven B[a]P reduction.
- Assessment of PAHs exposure for active population including mobility, commuting and working locations to account for complex variability of population density and consequent exposure.

As for health effect assessment a number of enhancements can be considered:

- Longer time series of PAHs exposure to support both short and long term analysis;
- Better accuracy in the determination of PAHs exposure;
- Higher space resolution of PAHs exposure to account for street level effects and urban hotspots.

4. Level of portability of employed methodology

To reproduce these methodologies to other geographic contexts a high degree of portability should be available. The design of the EXPAH project was planned to have a demonstration character, using the city of Rome as test area. The methodologies developed during this experience have some degree of portability in different urban contexts, provided that city specific keys data (both environmental and health data) are available. Hereafter the portability of EXPAH methodology is discussed in detail.

Level of portability of EXPAH methodology: The EXPAH project created an unforeseen compilation of standard methodologies for the assessment of population exposures to PAH compounds including air quality monitoring, modeling, population time activity, assessment of infiltration, and source identification. As a result the annual and spatial patterns of PAH exposures of children and the elderly were characterized in detail, allowing for development of efficient risk reduction policies. These results were published in a number of peer-review journals and several scientific conferences.

As far as the monitoring, modeling and health effects methodologies are concerned, it has to be considered that their application requires very high multidiscipline skills, normally available in specific research groups. Out of this scientific context, it would be difficult to find the required know-how to implement the proposed methodologies. So, in order to export them in urban contexts without sufficient skills, support would be necessary to gather the needed data and to apply the EXPAH methodologies. To improve the level of portability, the PAHs monitoring activities were carried out by using standard and consolidated methods for sampling and chemical analysis. The employed techniques are easily replicable at the scientific or technical departments of environmental agencies.

Some of EXPAH's results and deliverables can be reutilized in a different geographic area, as they can be considered representative of a general urban Mediterranean context. In this way some of the expensive and time consuming surveys and sampling activities do not need to be replicated, improving the level of portability. Among them we can find:

- Time activity data of children and elderly people living in Rome;
- PAHs infiltration factors for home, school, office, car, bus;
- 2020 PAHs National emissions in the 2020CLE and 2020 AME scenarios.

Level of portability in other urban areas or mixed industrial/urban areas: The EXPAH project was implemented in the city of Rome, as it was considered as representative of a large Mediterranean urban area. Its methodologies to assess PAHs population exposure can be exported in other metropolitan areas or medium size cities eventually with a mixed industrial/urban context, provided the expertise is available. The mixed contexts of particular interest due to the complains about health effects produced by large industrial facilities, such as steel plants, which are often located close to urban and harbor areas. In such a context with high exposure levels, it might be important to assess PAHs exposure, source contributions and to identify mitigation scenarios.

The EXPAH project has provided methodologies that are able to answer to the main questions existing in such contexts. Unfortunately, they are not completely portable unless some adaptations to the specific situation are planned. Among them we find:

- Seasonal PAHs monitoring at outdoor level to provide actual PAHs exposure of population and data to confirm the model performance;
- Identifying the main sources including the contribute from biomass combustion;
- Developing PAHs, PM2.5 and gaseous pollutants emission inventory;
- Making available cartographic data of the studied region; Population density data; Health data of mortality and morbidity.

A large fraction of the local information mentioned is usually available in the European countries.

Other important data- such as population time activity and PAHs infiltration factors, can be either exported from EXPAH results or derived locally using the EXPAH methodologies.

As far as the above data are available and skilled or supported expertise in AQ modeling and epidemiology are accessible, the application of methodologies described in Radice et al. (2012), Silibello et al. (2013, 2014) and Cesaroni et al. (2014), should provide results on PAHs population exposure for the studied region.

Based on the above assumptions, a medium level of portability of the EXPAH's methodologies can be proposed.

5. Conclusions and remarks

The EXPAH project provided a set of methodologies to get information on PAHs exposure of population living in large metropolitan areas. It was possible to assess the actual PAHs exposure in the most living environments in different seasons, to estimate daily PAHs exposure at urban level with a spatial resolution of 1 Km^2 and to evaluate health outcomes due to PAHs exposure as well as the future impacts on exposure and health to the 2020 expected levels. The employed methodologies have revealed a medium degree of portability in other urban areas. Although the sampling and analysis techniques for PAHs monitoring were kept at standard level to easily transfer them to environmental authorities, the remaining modeling and epidemiologic methods need specific context data to be applicable in other areas. Other EXPAH results, such as PAHs infiltration factors and population time activity data can be considered portable in other urban Mediterranean areas. Finally, as EXPAH methodologies were developed in a scientific context, their applicability is strongly dependent on the skill of persons involved in transferring methods and data. External support would be needed to achieve full portability.

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