EXPAH - ACTION 4.1: Collection of raw emission inventories and their upgrading

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Riferimento
ARIANET R2011.13

May 2011

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1 Introduction

The aim of EXPAH project is to describe population exposure to Polycyclic Aromatic Hydrocarbons (PAHs) compounds content in particulate matter and to quantify the environmental and health problems induced by these emissions. PAHs are produced by incomplete combustion of organic matter and are emitted by five major sources: domestic heating, vehicles exhaust gases, industrial combustion (including power generation and waste processing), agricultural and natural biomass burning. In urbanized areas domestic heating is usually the largest contributor of PAHs, even if Rome can receive contributions from mobile sources, two large airports (Fiumicino Hub and Ciampino), a urban waste landfill, two waste incinerators and an oil refinery located within the city suburbs.

In order to develop a complete emission inventory for the city of Rome and its surrounding area, it has been necessary to integrate different source of information characterized by diverse scales and space resolution, identify inconsistencies and integrate them to cover the space areas that will be covered by air quality model simulations.

Since we found out that Lazio Region emission inventory does not include PAHs, the Italian national Emission inventory (ISPRA2005), and other inventories available at European level (i.e. EMEP, TNO) have been used in the integration process. The national inventory is based on top-down methodology and it has been disaggregated at province level. European inventories are distributed as gridded pollutants emission that are therefore not directly connected with administrative units.

In this report results of this integrated approach are presented.

2 Emission Inventories

The most important dataset we referred to it is the National Emission Inventory (hereafter ISPRA2005) which was prepared by ISPRA (former APAT) using a top down approach (ISPRA, 2009); in the same way provincial (NUTS 3 level) estimates have been disaggregated to a municipal level (NUTS 4), adopting activity data. The municipal emission inventory (INCOM05) is the starting point for the creation of the input to the model.

At the national level emission inventories are compiled thanks to the use of emission of pollutant per unit of activity (Emission Factor, EF), derived, for each kind of source, from studies, articles and international reports. In order to give the annual estimated emission for specific sector EF are multiplied for that sector activity.

Table 1. Total PAH emissions (kg/year) in Lazio Region (source: ISPRA2005).

	LAZIO REGION	ROMA (province)	ROMA (municipality)
Energy Production	52	26	3
Comb in Residential	4389	3086	2010
Comb.in Industry	2	1	0
Prod. Processes	19	19	6
Solvent Use	0.5	0.3	0.1
Road Transport	176	114	86
Other Transport & Mobile Machinery	29	13	6
Waste Treatment	1310	334	64
TOTAL	**5977**	**3593**	**2175**

In the Italian Emission Inventory, PAHs are estimated in their complex, without splitting among the different congeners. A summary of emissions for Lazio Region is given in Table 1 and Figure 2.

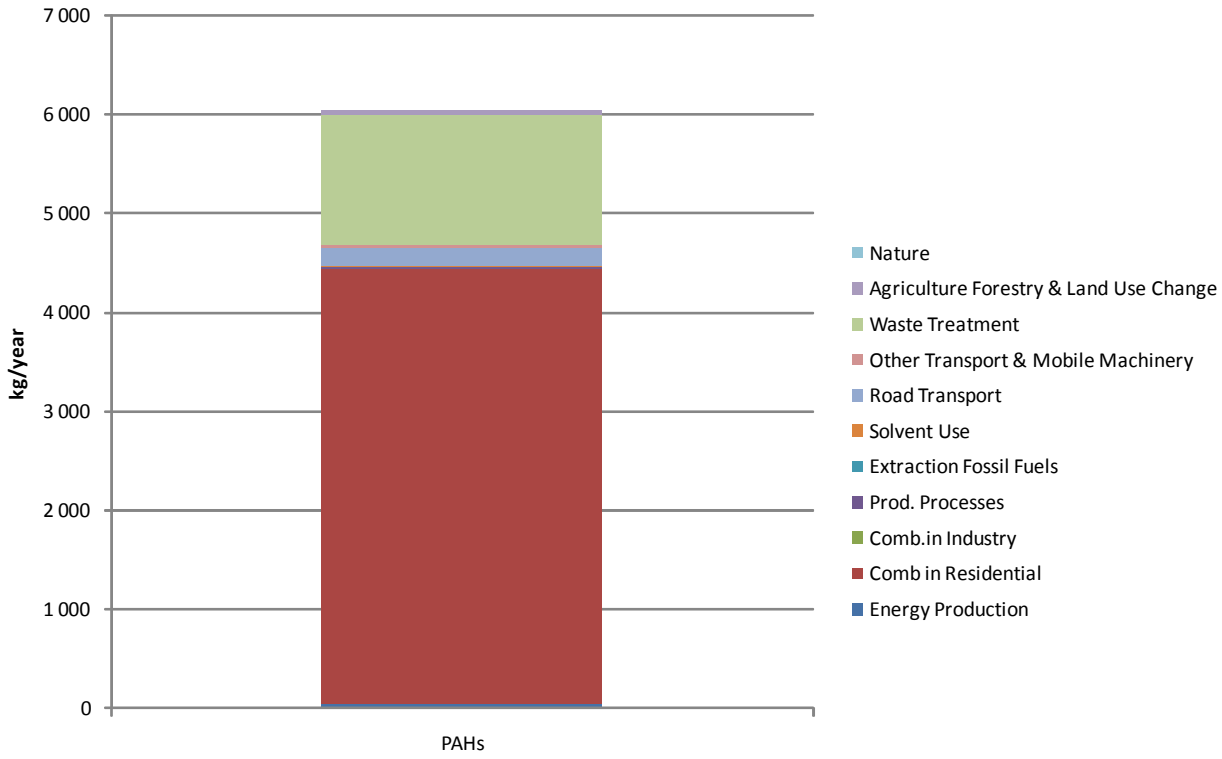

Figure 2. Total PAH emissions in Lazio Region (source: ISPRA2005).

To give a better understanding of environmental problems and to reduce environmental impacts, it's important to identify major pollutant sources. For Lazio Region it's possible to say that PAHs are produced, first of all, by domestic heating and waste processing, and then by incomplete combustion in vehicle exhaust gases, industrial combustion (including power generation), agricultural and natural biomass burning.

The geographic distribution (Figure 3) of diffuse emissions show that, by using local proxy variables, it's possible to locate different sources in the considered area. Residential heating. is by

far the major source accounting for 73% of emissions on Lazio Region, that grow to 86% on Rome Province and 92% on Rome city. Waste treatment contribution is the second main contribution with 22% of emissions over the Regions, 9% over the province and 3% over Rome municipality. Road transport contribution remains in the 3-4% range over the different considered areas.

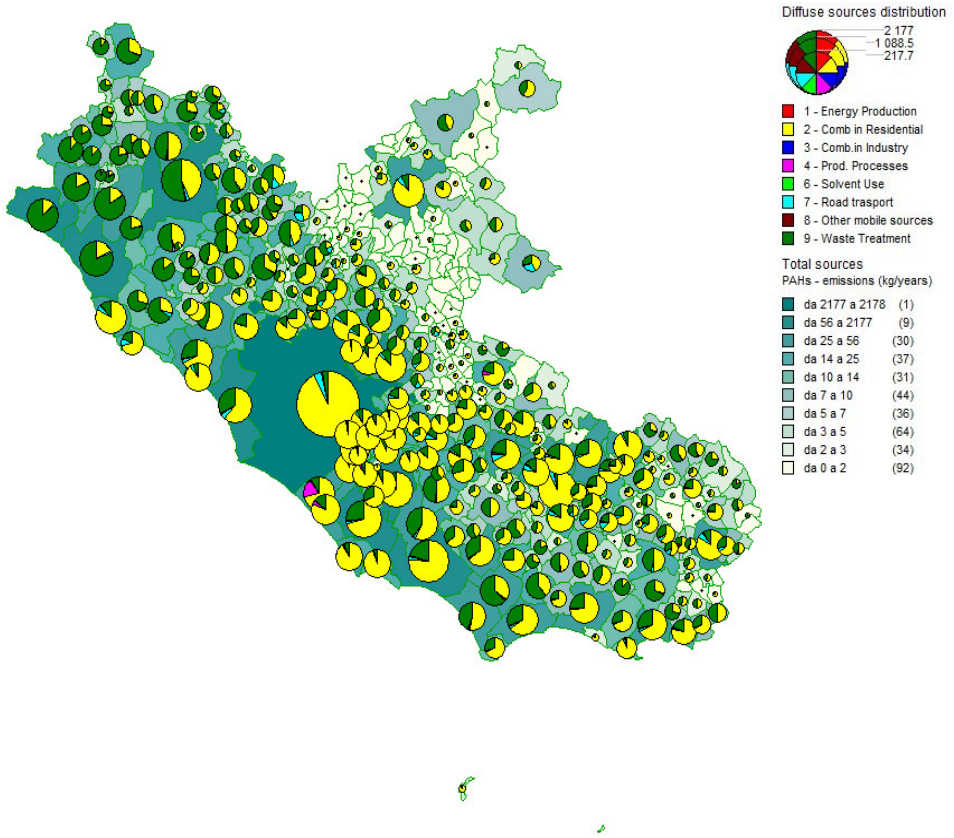

Figure 3. Total PAH emission distribution in Lazio Region (source: INCOM05).

Using historical trends of total national PAH emissions (Figure 4), it will be possible to update emission inventory data, which are referred to year 2005, to the target year of EXPAH field campaigns and air quality modeling activities (2011).

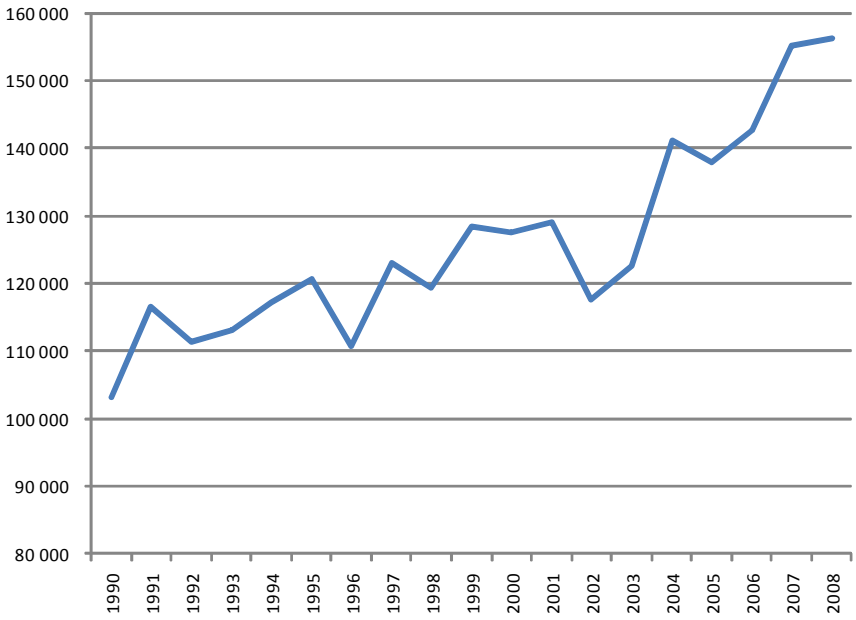

Figure 4. Total PAH national emission trend (source: ISPRA).

3 Speciated PAH inventory for EXPAH project

According to the International Agency for Research on Cancer (IARC), it's possible to identify a group of 6 PAHs as probable or possible human carcinogens, but in normal practices, and in particular in Italian Emission Inventory, PAHs considered are four, and are the same used as indicators for the purposes of emissions inventories under the United Nations Economic Commission for Europe (UNECE) Persistent Organic Pollutants (POPs) Protocol. The Protocol specifies that the following 4 PAHs should be used as indicators for emission inventories:

- benzo[b]fluoranthene
- benzo[k]fluoranthene
- benzo[a]pyrene
- indeno[123-cd]pyrene

Information about PAH speciation are not always available, and in some cases it's necessary to scale emission factors from a single measurement to all the sectors. For example it could be useful to assume that the species profile of all the plant of the same activity sector are similar.
Emission Factor data used in this project to speciate total PAHs present in the Italian Emission Inventory include research reports, journal articles and contact with industry; in some case different source of information as Best Available Technology (BAT) reports, and direct measurements of industrial stacks have been considered.
The most important sources used to realized the PAHs speciation are the Emission Inventory Guidebook (EMEP/EEA 2009), COPERT III methodology (Kouridis et al., 2000) and, for natural mass burning, some specific measurement (Finlayson-Pitts and Pitts, 1999).
For this reason inventory speciation is subject to a certain amount of uncertainty and it has been useful to compare our results to emission data coming from TNO European database and already subdivided among the mentioned 4 congeners.

In Figure 5 and Figure 6 some examples are presented; it's important to stress how different can be speciation profile also in the same emission category (Energy Production and Residential Heating), often the driver is the type of fuel that can originate very different profiles (Figure 6).

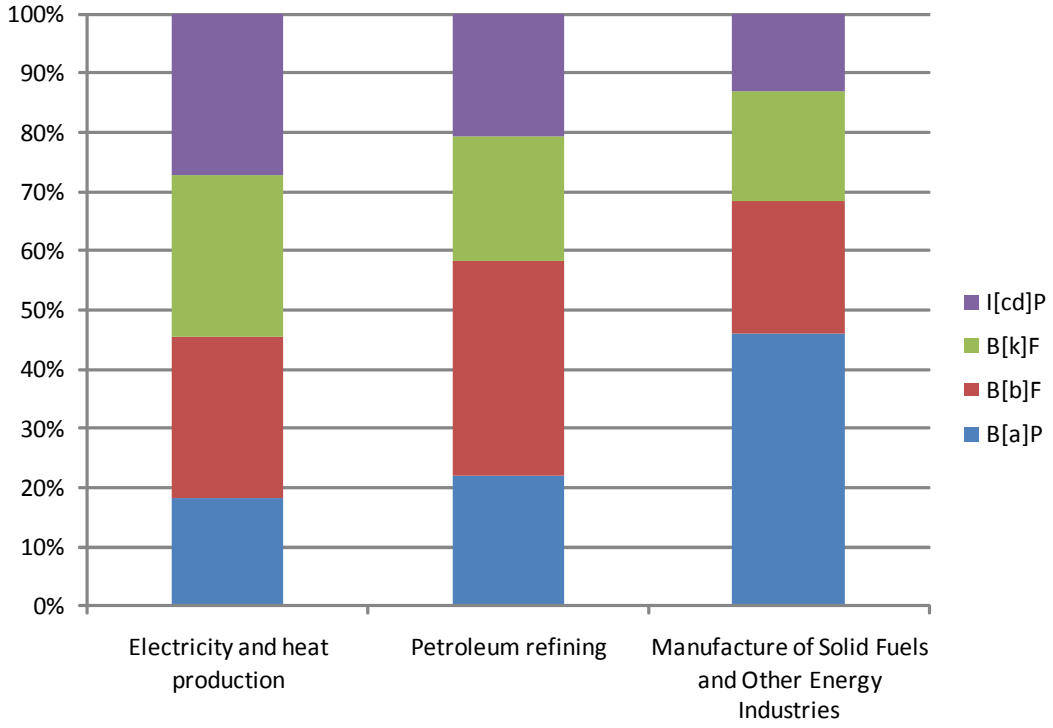

Figure 5. Contribution of different congeners in the Energy Production Emission (source: Emission Inventory Guidebook 2009)

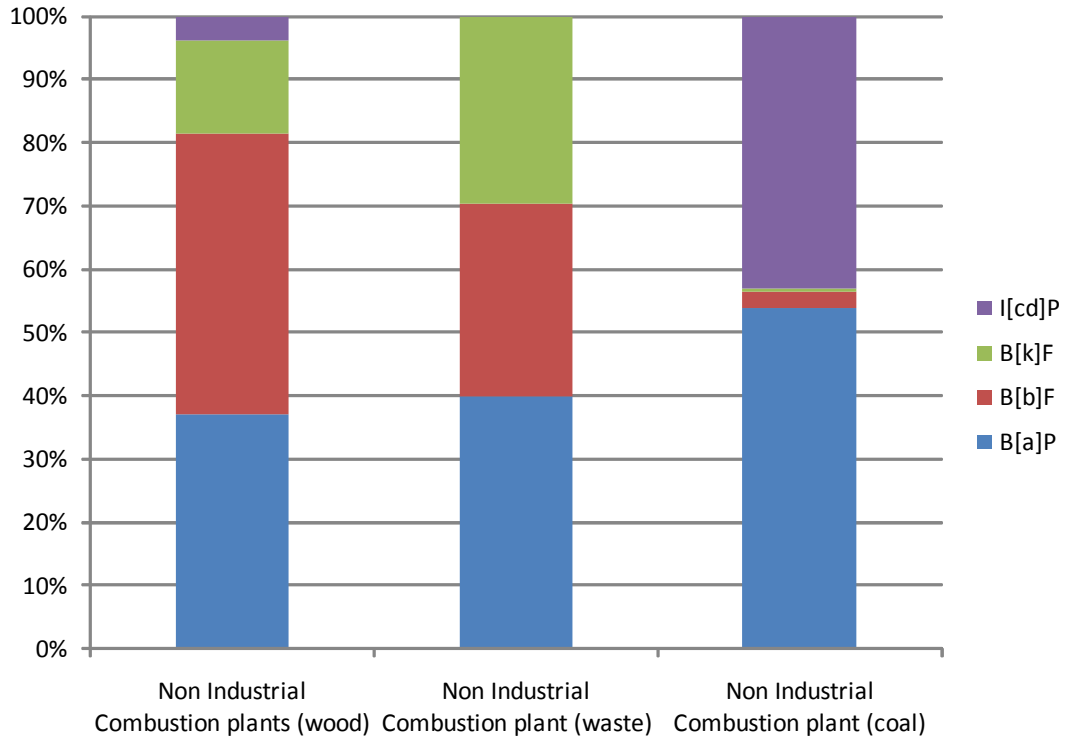

Figure 6. Contribution of different congeners in Non residential Combustion Plants (source: Emission Inventory Guidebook 1999)

Using different profiles it has been possible to speciate all PAH Lazio Region emissions and results are briefly resumed in Figure 7 and Figure 8.

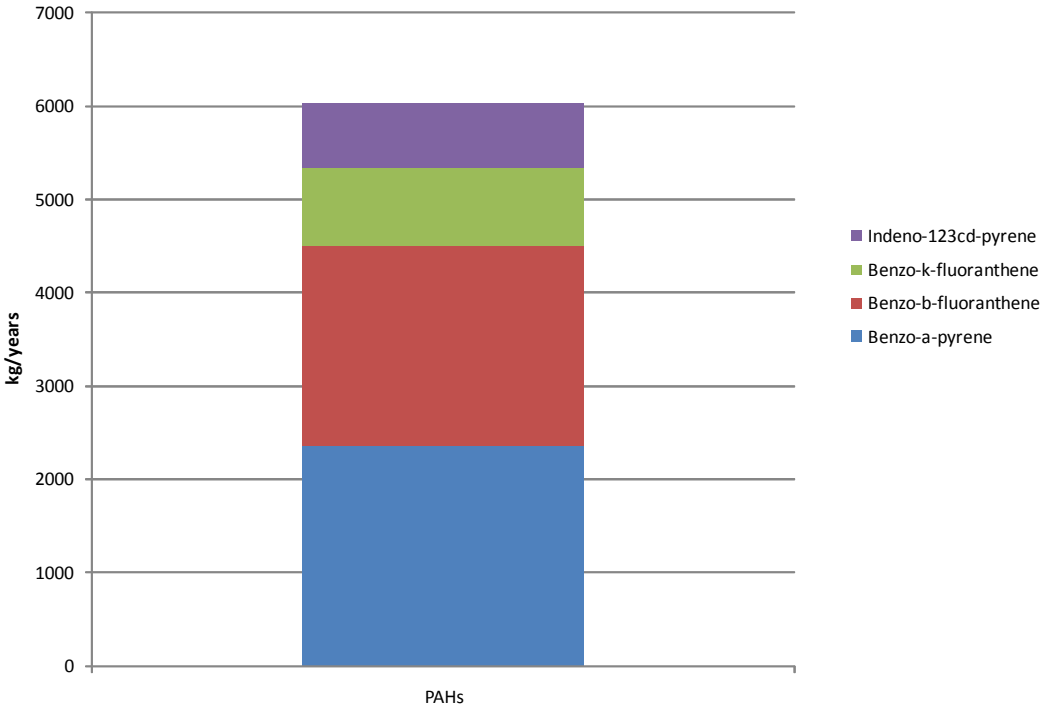

Figure 7. Speciated PAH emission in Lazio Region.

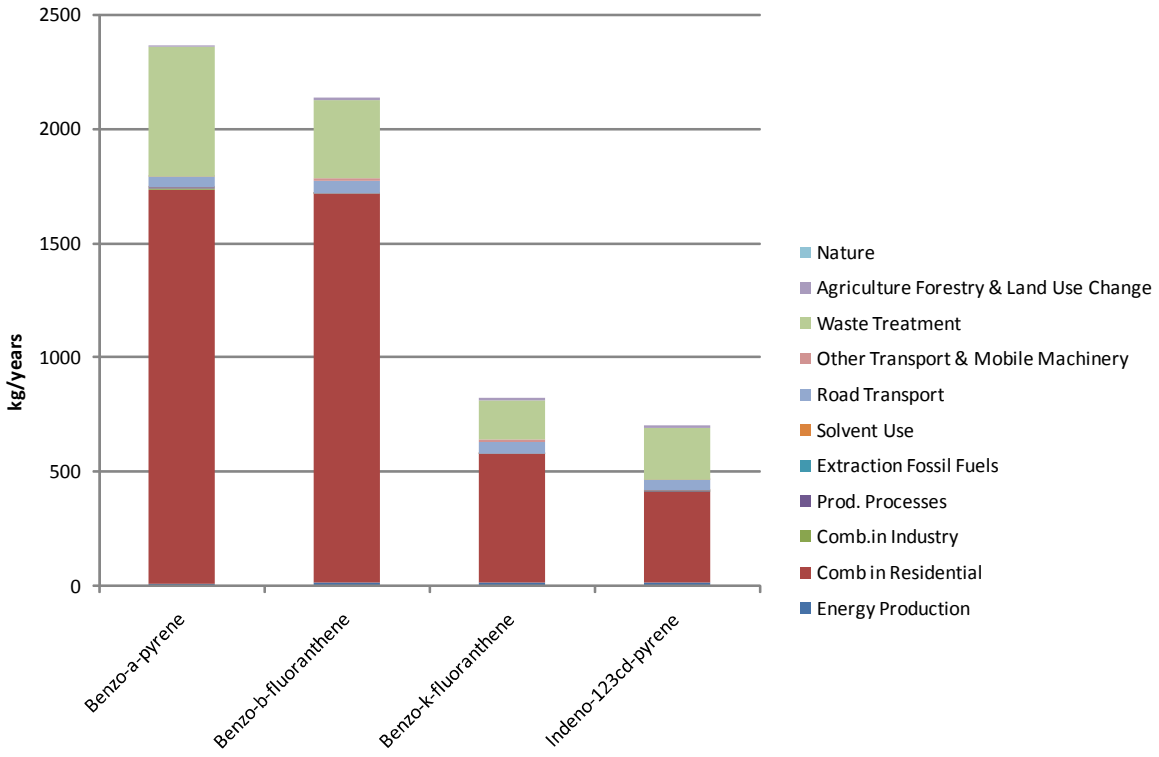

Figure 8. Speciated PAH emission in Lazio Region (sector contributions)

In order to better understand results of the speciation, it's interesting to compare them with those provided by TNO.

TNO made an emission inventory for persistent organic pollutants (POP) for the year 2000 based on submissions of emission data from the Parties to the Convention on LRTAP (Denier van der Gon et al., 2007). This database covers all the UNECE countries, except United States and Canada, and uses default emission estimates to complete the inventory where sources or compounds lack in official submissions.

A first comparison of TNO and national speciated PAHs emissions (ISPRA) is shown in Figure 9 and Figure 10. Data refer to year 2000, that is the starting reference year for TNO inventory.

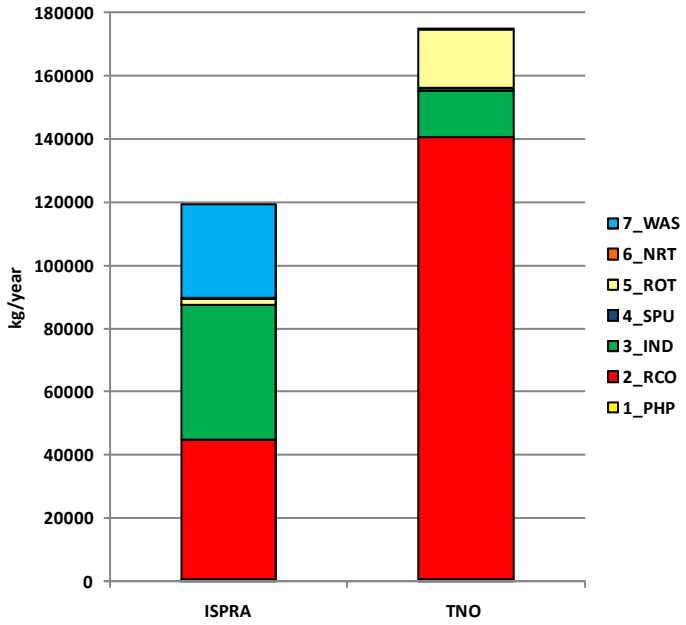

Figure 9. Speciated PAH emissions (sector contributions) (year: 2000)

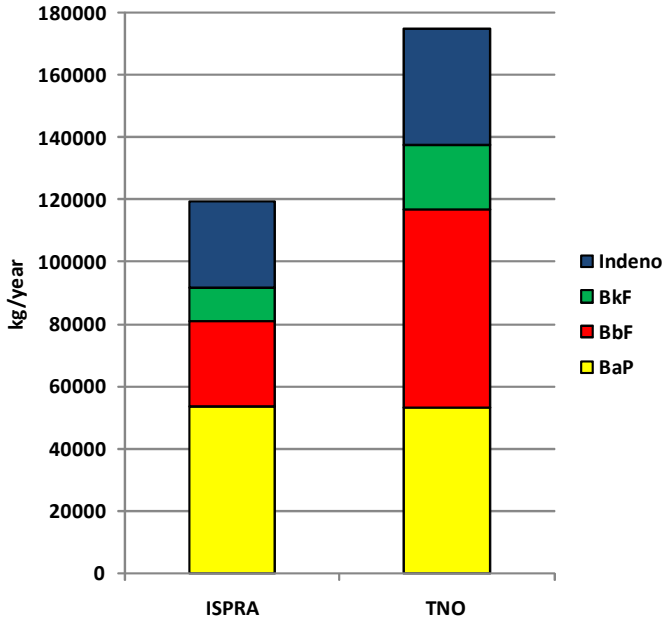

Figure 10. Speciated PAH emissions (congeners contributions) (year: 2000)

It is possible to observe that the total emissions are quite different, TNO inventory reports values 46% higher than the ISPRA ones, first of all due to the big difference between estimations of residential combustion sector (RCO), maybe related to different estimation of activities (fuel sold,..).

ISPRA industries emissions are bigger than TNO ones, and also in this situation it's very difficult to understand if this is produced by different factor activities or, probably, by different technologies.

Another interesting difference is the lack in TNO inventory of PAHs produced by waste treatment that in ISPRA represent 25% of total over the whole Italy.

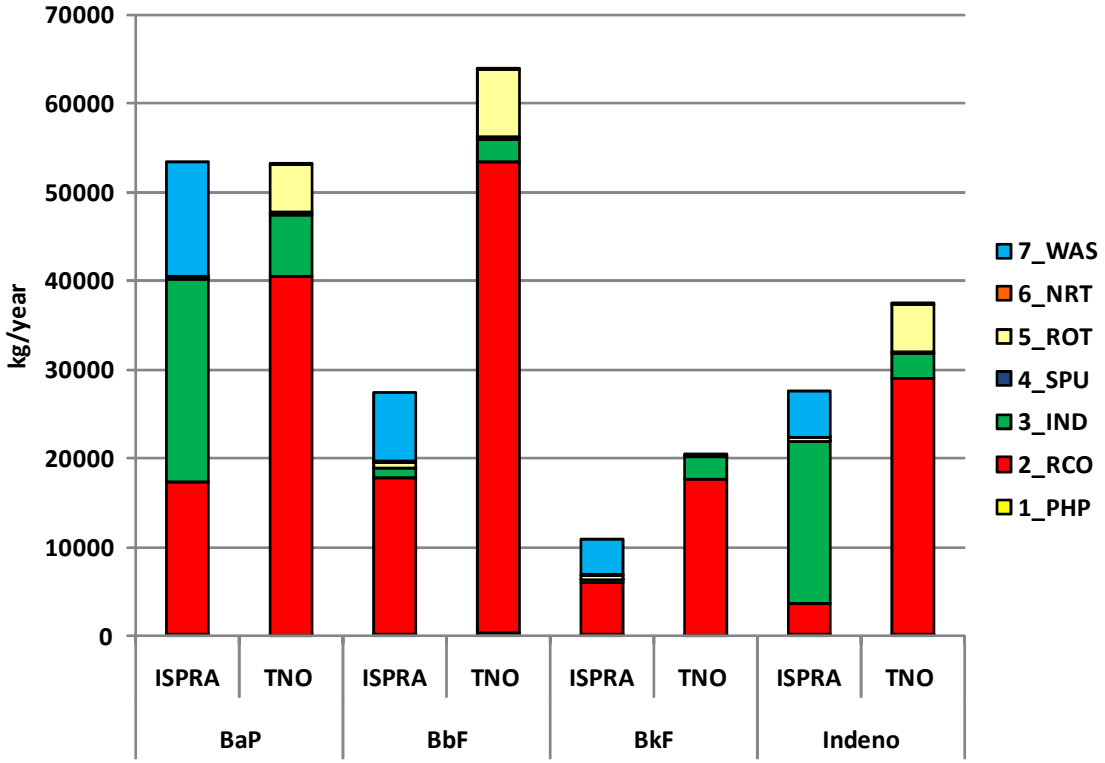

Figure 11. Speciated PAH emission (sector contributions) (year: 2000)

Figure 11 reports contributions of different sectors to the 4 considered congeners emissions. The total amount of BaP is practically the same for the two data sets, even if contributions from emission macro-sectors are different.
In TNO inventory, there is no BkF emitted by road transport, while in COPERT III its emissions are present, mainly produced by heavy duty vehicles.
The higher Benzo[b]fluoranthene emissions estimates by TNO compared to ISPRA ones could be due to a higher BbF emission factor for wood combustion or to a higher activity factor, or to a different type of fuel.

Starting from 2000 TNO European emissions, it's possible to extend the same databases comparison to year 2005. After establishing 2000 reference year emission inventory, TNO built the projections to future years (2010, 2015 and 2020) assuming that current legislation and existing protocols full implementation needs to include a preliminary inventory of possible additional reduction measures (Denier van der Gon et al., 2007). The considered future scenario is based, as much as possible, on the baseline scenarios developed in the framework of the Clean Air for Europe (CAFE) program (Amann et al., 2005).
To define TNO 2005 emissions it's been necessary to interpolate 2000 reference year with 2010 (scenario) values. Results are shown in Figure 12. For year 2005 the two datasets absolute differences are limited (about 10%) but contributions from different PAH congeners remains quite different.

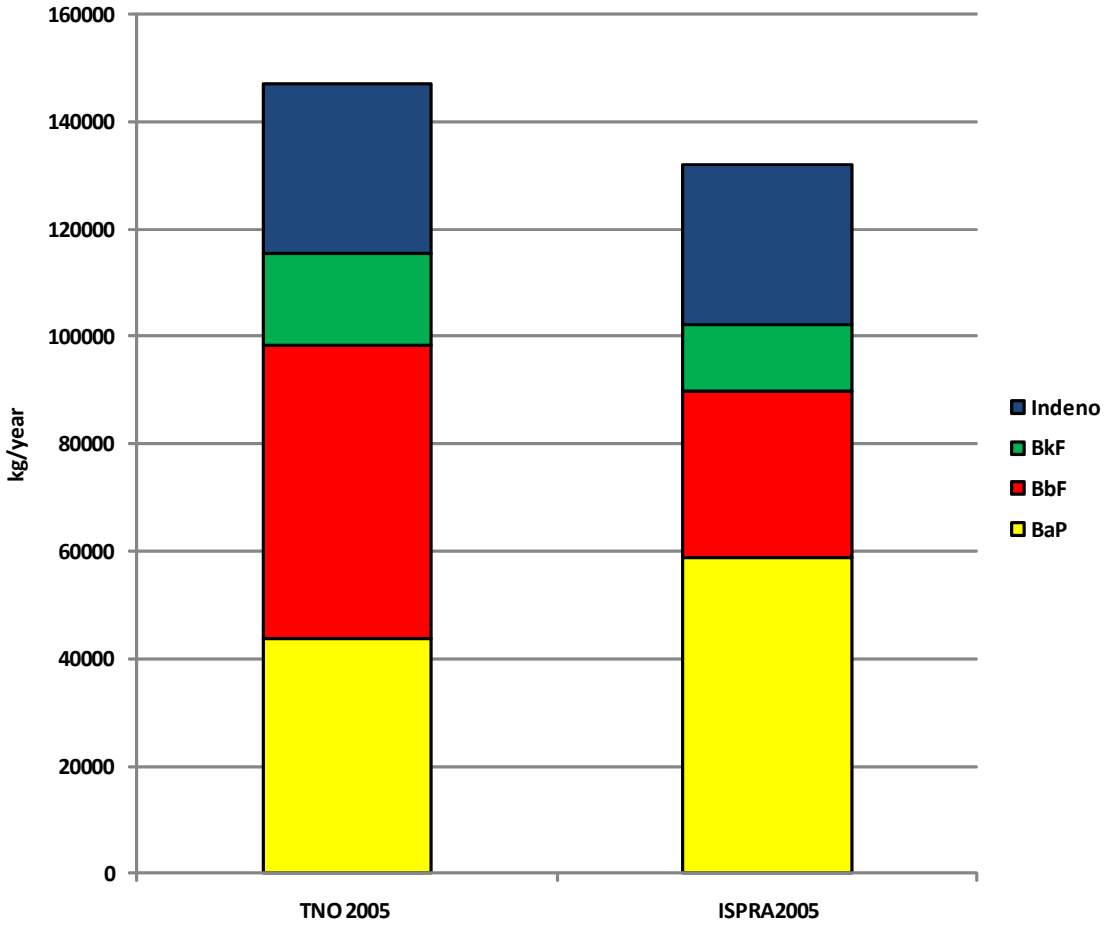

Figure 12. Speciated PAH emissions (congeners contributions) (year:2005)

The brief comparison of the available European and national scale emission datasets evidences that PAHs emission estimation is affected by a large uncertainty. Highlighting these uncertainties is necessary for the consequent evaluation of the air quality and health impact evaluation of PAHs emissions. Moreover it rises the need to improve emission inventories and to extend their technical contents to include more chemical species and possibly the description of the main emission sectors profiles uses to derive the values provided.

4 Conclusions

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 the possibility to build emissions characterized by high space resolution over the target area of Rome and the surrounding region. 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 PAHs 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.
Results obtained have been analyzed and compared with the European scale emission inventory developed and provided by TNO. Differences have been highlighted and possible causes discussed. TNO emission inventory will be used as the background reference dataset to cover areas possible not included in the national inventory.
The different datasets emission intercomparison set in evidence the large degree of uncertainty that affects PAHs emissions and that can generally be considered larger than that associated to other pollutants.
The analysis of emissions over Lazio Region and Rome metropolitan area confirmed that the main sources of PAH are, in order of relevance:

- Combustion in residential heating,
- Waste treatment,
- Road transport;

With the first sector accounting for 73% of emissions in Lazio Region growing to 92% within Rome municipality.

The emission inventories analyzed and elaborated, together with the emissions computed from the evaluation of traffic flow over Rome street network, will be the basis to build gridded hourly emissions for the whole year 2011.

5 Aknowledgents

We thank TNO for kindly providing European scale PAHs emission inventory for its possible use within EXPAH project. We would like to thank in particular Dr. Hugo Denier van den Gon for his helpfulness and for the fruitful discussions about PAHs emission estimate methods.

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