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Indoor and Outdoor Air Concentrations and Personal Exposure for Selected Hazardous Air Pollutants (HAPs) in European Cities

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RESEARCH ARTICLE

ABSTRACT

Over the decades, the attention of atmospheric scientists and air quality managers focused on the quality of outdoor air and its impact on human health. Much data is now available on indoor and outdoor air quality in Europe, but very little convincing information exists about people's exposure to air pollutants. Nonetheless, human exposure to air pollutants occurs mainly indoors because people spend much of their time (on average 85 to 90%) in confined spaces (homes, office buildings, schools etc.). They are exposed to a complex mixture of air contaminants at concentrations often several times higher than outdoors.

While the quality of the data within each of the different studies carried out in Europe through QA/QC procedures is assured, the comparability is highly questionable for the data provided by the measuring campaigns because various methodological approaches applied to get analytical data for the concentration of pollutants and methods used for exposure assessment. Moreover, measuring campaigns carried out at different seasons and climatic conditions make it rather challenging to obtain an objective outcome on human exposure to hazardous air pollutants and evaluate the overall risk to human health.

Analysing the results of the measuring campaigns conducted in Europe is quite evident in need to develop and adopt harmonised measurement methods and protocols and exposure guidelines considering various factors affecting the quality and representability of the measurements. The paper provides information on indoor/outdoor air concentrations and personal exposure monitoring for two priority pollutants, i.e. benzene and formaldehyde, in European cities.

KEYWORDS

Exposure, Indoor/Outdoor, Benzene, Formaldehyde

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INTRODUCTION

The current European Union (EU) policy and plans on urban air quality include that citizens are to be effectively protected from risks to health from air pollutants. The EU legislation addresses air quality standards, national emission ceilings and emissions from vehicles and industries. Given the time most people spend indoors (ca. 85-90%), human exposure to hazardous air pollutants is governed to a great extent by the concentration of indoor pollutants rather than by ambient air concentrations.

Hazardous air pollutants (HAP) have many potential sources inside residential (homes) and workplaces (office buildings, schools). Due to the accumulation of contaminants in confined spaces, indoor air pollutant concentrations might be up to several times higher than outdoor air concentrations. The 6th Environmental Action Plan of the European Commission (2001- 2010), taking into consideration the existing knowledge on possible health risks associated with the presence of HAPs in non-industrial indoor environments, including for the first time as a priority, research on indoor air quality with an emphasis in quantifying and reducing the overall exposure of the European citizens to hazardous air pollutants.

In this context, EU-funded research projects were carried out with the aim to identify and quantify the main determinants of air pollution in different indoor environments (homes, office buildings, schools), map them and explain their distribution at the European level and evaluate human exposure to HAP. A selection of these projects is listed below:

EC-AUDIT	THADE	RADPAR
EXPOLIS	EPHECT	
MACBETH	HEALTHVENT	
PEOPLE	OFFICAIR	
INDEX	SINPHONIE	
AIRMEX	HESE	
BUMA	SEARCH	
ENVIE/IAQIA	HITEA	

The knowledge gained in the frame of these projects could also be of interest for assessing the local and regional impact of climate change (e.g. heat waves, excessive precipitation, humidity and temperature changes) on indoor environmental quality.

In 2005, the INDEX project (Critical Appraisal of the Setting and Implementation of Indoor Exposure Limits in the EU) was financially supported by EC-DG Consumer Protection (SANCO) and the JRC (the Commission's Joint Research Centre, Ispra, Italy) and carried out in collaboration with renowned scientific institutions in Europe presented a first approach in assessing and regulating the presence of priority substances in indoor air [1].

In the frame of the INDEX project, the existing knowledge worldwide has been assessed on

- Type and levels of chemicals in indoor air and the
- Available toxicological information to assess risk to health and comfort.

In 2010, WHO (World Health Organisation), published the Guidelines for Indoor Air Quality (selected pollutants)-"Acknowledging that indoor air has a special role as a health determinant and that the management of indoor air quality requires approaches different from those used in outdoor air" (WHO Guidelines for indoor air quality) [2]. According to WHO, "the guidelines for indoor air quality provide a scientific basis for legally enforceable standards". The Guidelines targeted public health professionals and might serve as well for authorities involved in building design and use of indoor materials and products.

Indoor air quality is becoming an emerging environmental health issue and is strongly connected with topics related to the energy performance of buildings, the quality of the construction and building materials and the behavioural activities of the citizens.

Relevant EU-Directives/ and Guidelines for building performance and indoor air quality are:

- The Energy Performance of Buildings Directive [91/2002/CEE], [The recast Directive on Energy Performance of Buildings (EPBD) 31/2010]
- The Construction Products Regulation (CPR) [305/2011/CEE]
- The EU Green Paper on Smoking Ban (EU-COM 2007)

During the last decades, a few studies (EXPOLIS, 1996-1998; MACBETH, 1997-1998; AIRMEX study, 2003-2008; SINPHONIE, 2011-2012) carried out at pan-European level to gather information on the occurrence of air toxics in different environments (indoor/outdoor), to evaluate the exposure of the European population to air pollutants and to identify potential sources, in particular, in residential places, public buildings and schools [3-6]. Apart from EXPOLIS, MACBETH, AIRMEX and SINPHONIE, measuring campaigns (German Environmental-GerES IV- study, 2001-2002; Duarte-Davidson et a.,1999; Crump et al.; Levsen et al.; Gonzalez-Flesca et al.) conducted at the local level by national research institutions and local authorities providing data for health risk evaluations in support of EU and State regulations [7-11] .

Despite the large amount of data now available on indoor and outdoor air quality in Europe, very little convincing information exists about people's actual exposure to air pollutants.

While the quality of the data within each of the studies mentioned above, through QA/QC procedures, is assured, the comparability is highly questionable for the data provided by the different campaigns because various methodological approaches applied to get analytical data for the concentration of pollutants and methods used for exposure assessment. Moreover, measuring campaigns carried out at different seasons and climatic conditions make it rather challenging to obtain an objective outcome on human exposure to hazardous air pollutants and evaluate the overall risk to health.

The paper provides information on indoor/outdoor air concentrations and personal exposure monitoring for two priority pollutants, i.e. benzene and formaldehyde, in European cities. The aim is to make suggestions on the design and execution of future measuring campaigns based on harmonized measurement methods and protocols, and exposure guidelines for risk assessment.

MATERIALS & METHODS

The methods and techniques used for the different measuring campaigns significantly differ from one study to another. In the EXPOLIS study, sampling for volatile compounds (including benzene) was made using personal pumps for 48 hours, while in the MACBETH study, the diffusion technique (passive sampling) was used over five days. A similar methodological approach was applied during the measuring campaigns in the AIRMEX and SINPHONIE study frame. In other studies, sampling using personal pumps was done four times per day in weekly measuring campaigns timely located in the four seasons of the year (Lahaniati et al.) [12].

The lack of a standard procedure regarding the representativeness of sampling location, measuring time, micro-environmental activities and the subset of the population leads to a considerable diversification of the data. Finally, it results in exposure estimates and assessments of indicative value. An example is the city of Athens, where three different values for the outdoor concentration of benzene are reported, although the measuring campaigns were carried out in almost the same period (between 1996-1999). In work by Lahaniati et al., the mean ambient air concentration for benzene in Athens/Greece was 32.5 µg/m³ while the corresponding values within the EXPOLIS and MACBETH studies were 11 and 20.7 µg/m³, respectively. Since large-scale measuring campaigns are costly, time and personnel intensive, it would be reasonable to apply generally accepted procedures for measuring HAP and assessing population exposure to air pollutants. One way could be to base personal exposure assessments on data obtained over an extended period (one year-as monthly average values), considering micro-environmental activity patterns, taking into account the overall behaviour of individuals during rest and working hours. In specific cases, when the impact of construction materials, paints, furniture and specific behavioural activities on the overall indoor air quality are evaluated, additional targeted measurements should be performed.

Setting up limit values indoors would significantly improve the quality of construction and building materials and other products to be used in indoor environments and will indirectly increase public awareness [1,2,13] . This would also fit well with the Integrated Product Policy (IPP) concept on the environmental and health impact of materials and products during their entire life cycle.

INDOOR/OUTDOOR AIR CONCENTRATIONS, EXPOSURE ESTIMATES

BENZENE

In the frame of EXPOLIS, MACBETH, AIRMEX and SINPHONIE studies in the last decades, extensive measuring campaigns have been carried out at the pan-European level to measure indoor/outdoor concentrations for benzene (among other pollutants), which IARC classifies as known human carcinogen (Group 1) and to relate them to personal exposure estimates. The results indicate that ambient air concentrations for benzene substantially vary between the northern and the southern part of Europe, with higher ambient air levels measured in the cities of Southern Europe. It is mainly due to climatic conditions (higher temperature, low wind speed regimes), heavy traffic and often the lack of infrastructure needed to facilitate the movement of the citizens from/ into the city. While for Athens, Murcia, and Milan, outdoor (mean) concentrations up to 21 µg/m³ were measured, in Copenhagen, Helsinki, Rouen and Prague, outdoor concentrations for benzene reached values up to 5 µg/m³. In indoor environments (homes, public buildings and schools), mean benzene concentrations range from a low of 2.2 to a high of 13.2 µg/m³. Personal exposure monitoring generally shows higher concentrations than benzene concentrations indoors and outdoors. From the data available until now, it is evident that Northern European cities have higher exposure/outdoor (E/O) and indoor/outdoor (I/O) concentration ratios than cities in Southern Europe, ranging from 0.9- 3.7 and 0.5- 2.3 for E/O and I/O, respectively (table 1).

City	Personal	Indoor Air	Outdoor Air	I/O	E/O
Athens (1)	18	11	11	1	1.6
Basel	5.6	3.1	1.5	2	3.7
Helsinki	3.4	2.2	1.6	1.3	2.3
Milan	16	13.2	10	1.3	1.6
Prague	12	12	5.2	2.3	2.3
Athens (2)	18.8	10.1	20.7	0.5	0.9
Padua	10.6	7	8	1.3	1.3
Rouen	13.4	9	4.7	1.5	2.8
Copenhagen	6.6	4.5	3.1	2.3	2.1
Murcia	23.1	12.3	11.7	2	1.9
Antwerp	12.2	9.4	4.4	1.6	2.8
(1) EXPOLIS study (2) MACBETH study					

Table 1: Personal exposure, outdoor/indoor concentrations (in µg/m^3) and ratios exposure/outdoor (E/O) and indoor/outdoor (I/O) for benzene in different European cities.

In the frame of the German Environmental Survey of Children and Teenagers (GerES IV), Ullrich et al. reported on personal exposure (mean) concentrations for benzene of 2.5 µg/m^3, significantly lower than those obtained in cities of Central Europe (Prague, Antwerp). Hence, a daily intake of ca. 60 µg is estimated.

Gonzalez-Flesca et al. reported on personal exposure concentrations in the city of Nancy ranging from 9.9-55.5 µg/m^3, with a mean value of around 23.8 µg/m^3, which is significantly higher than the (mean) indoor and outdoor concentrations of 10.8 and 4.4 µg/m^3, respectively. Calculating the mean personal exposure concentration of 23.8 µg/m^3, a daily intake of up to 570 µg of benzene for 24 hours is estimated.

In the UK, ambient air concentrations of benzene are generally in the range of 1-6 µg/m^3. In homes, mean indoor air concentrations were estimated to be 8 µg/m^3. However, non-occupational exposed adults receive very high daily doses of 74-528 µg of benzene, which corresponds to an average range of benzene in air of 3.7-26.4 µg/m^3, an amount significantly higher than the mean outdoor air benzene concentration (Duarte-Davidson et al.). Crump et al. reported that the mean personal exposure for individuals in Hertfordshire, England was 183.9 µg/24h. However, using the mean outdoor air concentration near homes to predict human exposure, a value of 92.6µg/24 h has been obtained.

The results of two pan-European campaigns [EXPOLIS (1996-1998) and MACBETH (1998-1999] carried out in several European cities show personal exposure (mean) concentrations to benzene of 12.5 µg/m3, indoor/workplace concentrations of 8.9 µg/m^3 and outdoor concentrations of 7.4 µg/m^3, respectively.

Applying the micro-environment time activity data elaborated from questionnaires in the frame of the AIRMEX measuring campaign (2003-2008) for the EXPOLIS and MACBETH campaigns, i.e. 92% of the daily time spent in indoor environments (home+work) and 8% in outdoor environments [the reference daily population inhalation volume for adults amounts to 22 m^3, WHO ECH 170, Geneva, 1994], the contribution of the respective microenvironments to the measured (mean) inhalation exposure to benzene of 275 µg/day in Europe is estimated to be 179.7 µg/day (indoor/workplace) and 12.5 µg/day (outdoor), respectively.

In all measuring campaigns, there are differences between the daily inhalation exposure and the sum of the inhalation exposure measured in individual environments. In comparison, applying the same micro-environment activity data, the personal exposure (mean value) concentration measured during the AIRMEX campaign in eleven European cities in Southern-, Central- and Northern Europe is estimated to be 103.4 µg/day. The contributions of the respective microenvironments (home+work) and (outdoor) to the measured (mean) inhalation exposure to benzene in Europe are 67.7 µg/day (indoor/workplace) and 5.4 µg/day (outdoor), respectively (table 2) [13]. It indicates the existence of unknown or non-assessed sources to which humans are exposed to benzene during daily activities (active/passive smoking, un-foreseen/expected fuel emissions etc.).

One reason for the differences in the personal exposure concentrations of benzene between EXPOLIS/MACKBETH and AIRMEX is the reduction of the aromatic hydrocarbons in urban air due to the introduction of the automobile catalyst converter in the last two decades, the reduction of the benzene content of gasoline more recently and the ongoing reduction of toluene containing solvents in building and household products.

From all data available until now, personal exposure cannot be estimated from outdoor air concentrations only. Reducing benzene emissions from mobile sources will have a limited effect on total (indoor/outdoor) human air exposure to this compound; other benzene sources should not be underestimated. This observation has been made elsewhere too (Levsen et al.; Crump et al.; Gonzalez-Flesca et al.).

Benzene	Personal Exposure	Indoor/workplace	Outdoor	Day*
EXPOLIS/MACKBETH	12.5	8.9	7.4	275
AIRMEX	4.7	3.6	3.2	103.4
*Calculation based on the reference daily population inhalation volume for adults (22 m3) WHO ECH 170, Geneva, 1994				

Table 2: European micro-environmental and exposure concentrations (µg/m3) summarized from population based studies (EXPOLIS, MACKBETH, AIRMEX) [13].

FORMALDEHYDE

Formaldehyde (IARC) has been one of the most critical pollutants in indoor non-industrial environments. A large body of data exists on formaldehyde measurements in European homes and buildings. Indoor air concentration levels for formaldehyde range from a few µg/m³ up to 70 µg/m³, while mean outdoor concentrations of up to10 µg/m³ were measured. In air pollution episodes, formaldehyde concentrations can reach high values of up to 80 µg/m³ even at locations far from emission sources (Amanatidis et al.). However, in almost all measurements, formaldehyde indoor concentrations exceed by several times (5-20 times) the outdoor levels, indicating strong emission sources inside buildings and homes (Crump et al.; Gonzalez-Flesca et al.). According to a WHO study, exposure of humans to formaldehyde is mainly determined by its concentration indoors (ECA-EUR 13216 EN, 1990). A daily intake of 20 µg (HCHO) results from exposure to ambient air, while exposure to indoor and workplace concentrations has been estimated to amount to about 0.5-2 mg/day.

THE US

During the last 10-15 years, several studies were conducted in the US to determine indoor/outdoor air concentration levels for priority pollutants and assess personal exposure estimates. They have shown higher indoor than outdoor concentrations for the primary pollutants, especially for volatile organic compounds (US-EPA, Inside IAQ).

Indoor/outdoor (I/O) ratios, based on typical air concentration levels, of 2 and 50 µg/m³, for benzene and formaldehyde, respectively, are calculated—indoor (mean) concentrations for benzene range from 8.2 to17 µg/m³. "Typical values" for indoor and outdoor environments were up to 5 µg/m³. For formaldehyde, mean indoor concentrations reach 92 µg/m³, while "typical values" for outdoor air concentrations of 4 µg/m³ are reported. Daily exposure estimates are based on the assumption that people spend about 90% of their time in indoor environments and 10% outdoors. For benzene, daily personal exposures vary between 108-177 µg/m³.24 h. For formaldehyde, personal exposures range from 1080-2000 µg/24 h, similar to European exposure estimates.

CONCLUSION

The analysis of the existing data on indoor/outdoor air concentrations and exposure estimates for benzene and formaldehyde in European cities shows that indoor air concentration levels and the time spent indoors clearly determine human air exposure. However, there is a need to harmonise measuring protocols and techniques to ensure comparability of exposure data. Personal exposure monitoring over an extended period would be more appropriate than indoor/outdoor air concentration and individual exposure measurements during timely short measuring campaigns to obtain reliable human air exposure estimates. In this context, applying a proper sampling and analytical technique at low costs would be of high value. A comparison of the European and US studies' data indicates that indoor and outdoor air concentrations for benzene and formaldehyde are almost similar.

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