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EMISSION MODELLING OF HAZARDOUS AIR POLLUTANTS FROM ROAD TRANSPORT AT URBAN SCALE

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Abstract. This study is focused on the development of a modelling approach to quantify emissions of traffic-related hazardous air pollutants in urban areas considering complex road network and detailed data on transport activity. In this work a new version of the Transport Emission Model for line sources has been developed for hazardous pollutants (TREM-HAP). Emission factors for benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and also particulate matter (PM2.5) were implemented and the model was extended to integrate a probabilistic approach for the uncertainty quantification using Monte-Carlo technique. The methodology has been applied to estimate road traffic emissions in Porto Urban Area, Portugal. Hourly traffic counts provided by an automatic counting system were used to characterise the spatial and temporal variability of the number of vehicles, vehicle categories and average speed at different road segments. The data for two summer and two winter months were processed to obtain probability density functions of the input parameters required for the uncertainty analysis. For quantification of cold start excess emissions, Origin-Destination matrix for daily trips was used as additional input information. Daily emissions of hazardous air pollutants from road traffic were analysed for the study area. The uncertainty of the emission estimates related to the transport activity factors range from as small as −2 to +1.7% for acrolein and acetaldehyde on highways, to as large as −33 to +70% for 1,3-butadiene considering urban street driving. An important contribution of cold start emissions to the total daily values was estimated thus achieving 45% in case of benzene. The uncertainty in transport activity data on resulting urban emission inventory highlights the most important parameter and reveals different sensitivity of the emission quantification to the input data. The methodology presented in this work allows the development of emission inventories for hazardous air pollutants with high spatial and temporal resolution in complex urban areas required for air quality modelling and exposure studies and could be used as a decision support tool.

Keywords: road traffic emissions, hazardous air pollutants, air toxics, emission modelling, emission uncertainty.

1. Introduction

During the last decades, road traffic has become one of the most important sources of air pollution.

Among the extended number of chemicals emitted by the vehicles, hazardous air pollutants (HAP) require special attention due to their link with cancer and other serious adverse effects on human health. A list of 188 HAP, referred also as air toxics, was defined in Clean Air Act by the US Environmental Protection Agency (USEPA 2004) that contains pollutants associated with anthropogenic sources. Also, air toxics emitted by mobile sources, known as MSAT (mobile source air toxics) are identified, including: benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and diesel particulate matter (PM) (USEPA 2007). Emissions of MSAT are mainly related with incomplete combustion (e.g. benzene) and by-products formed during incomplete combustion (e.g. formaldehyde, acetaldehyde, and 1,3-butadiene), but evaporative processes of fuel components are also important. Besides, numerous measures to reduce air toxic emissions, including limits on gasoline volatility, limits on diesel sulphur, improvements in vehicle technology and performance, road transport is still one of the major sources of HAP especially in urban areas. Some studies indicate that mobile sources can contribute about 68% of total HAP emissions (Tam, Neumann 2004). Therefore, further studies to improve quantification of air toxic emissions induced by transport in urban areas where inhabitants are living close to the pollution sources are required to better cause-effect chain analysis.

Several methodologies to quantify road traffic emissions are currently available (e.g. Smit *et al.* 2007; Zallinger *et al.* 2005; Gkatzoflias *et al.* 2007). However, the modelling tools not always cover HAP or provide emissions with low temporal and spatial resolution that is not sufficient for urban scale studies. An intercomparison of the currently available models could be found at Barlow and Boulter (2009).

Urban emission inventories with higher temporal and spatial resolution are needed for a number of applications, such as urban air pollution modelling, population exposure modelling, definition of sustainable urban development policy, etc. The most commonly used technique to quantify the emissions is based upon the principle that the average emission factor for a certain pollutant and a given type of vehicles vary according to the average speed during a trip (Boulter *et al.* 2007a). For urban applications, hourly emissions for each road link are usually required. For this purpose, hourly traffic flows attributed to detailed road network that should be specified. Uncertainty of these data, as well as uncertainty associated with resulting emissions, is an important issue.

Quantitative methods for dealing with uncertainty in emission estimates involve the characterization of uncertainty in emission factors and/or activity data, and propagation of uncertainty to a total emission inventory. Although numerous probabilistic techniques have been applied for this purpose, the well-known Monte Carlo approach has multiple advantages and is the most often used for this purpose (e.g., Frey, Zheng 2002a, 2002b; Abdel-Aziz, Frey 2003). The IPCC (Intergovernmental Panel on Climate Change) and EPA have developed guidelines recommending the use of Monte Carlo methods as a part of a tiered approach for emissions uncertainty estimates addressing the quantification of uncertainty in emission and activity factors (IPCC 2000; USEPA 1997). Monte Carlo simulation methods are used to estimate uncertainty in inventories, such as for criteria pollutants, HAP, and greenhouse gases (e.g., Winiwarter, Rypdal 2001).

The present work intends to develop a modelling approach for quantification of traffic-related hazardous air pollutant emissions with high spatial and temporal resolution for the studies in urban areas. For this purpose, emission factors of HAP have been implemented into the Transport Emission Model for Line Sources (TREM). Also, this new version of the model was extended to integrate a probabilistic approach for the uncertainty quantification using Monte-Carlo technique. An application example of the developed methodology to the Porto Urban Area (Portugal) for the year 2008 is presented.

2. Methodology

2.1. TREM Emissions Model

The Transport Emission Model for Line Sources was firstly developed on the basis of COST319/MEET approach and focused on carbon monoxide, nitrogen oxides, Volatile Organic Compounds (VOC) including methane, carbon dioxide, sulphur dioxide and particulate matter with aerodynamic diameter less than or equal to 10 μm (PM10) (Tchepel 2003; Borrego *et al.* 2000, 2003, 2004). The prime objective of TREM is the estimation of road traffic emissions with high temporal and spatial resolution to be used in air quality modelling. Although the average-speed approach for the emission factors implemented in the model follows the European guidelines (EMEP/EEA 2010) the way how transport activity data are considered for the emission inventorying is conceptually different. Roads are considered as line sources and emissions induced by vehicles are estimated individually for each road segment considering detailed information on traffic flow provided by automatic counting system or from a transportation model. To process these data, TREM is directly linked to Geographical Information Systems (ArcGIS) and to the transportation model VISUM (Borrego *et al.* 2004).

A new version of TREM developed in this work use updated emission factors from ARTEMIS methodology (Boulter *et al.* 2007b; André, Joumard 2005). Following the definition of air toxics relevant for mobile sources, this new version TREM–HAP (Transport Emission Model for Hazardous Air Pollutants) is prepared to calculate the emissions of benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and also particulate matter with aerodynamic diameter less than or equal to 2.5 μm (PM2.5). The calculation algorithm is schematically represented in Fig. 1.

Firstly, exhaust hot emissions of total VOC, Methane (CH_4) and PM2.5 are estimated as a function of average speed for each class of vehicles. Both total emissions under thermally stabilised engine and additional cold-start emissions are considered due to the importance of cold-engine driving within urban areas. At next, methane hot emissions are subtracted from VOC and nonmethane VOC (NMVOC) emissions are separated into different compounds, including hazardous pollutants, using %-fractions as proposed by EMEP/EEA (2010) guidelines. MSAT cold start emissions are estimated as a function of average speed and ambient temperature. In this case, passenger cars only are considered due to the methodology limitations. An example of hot exhaust emission factors calculated for benzene and formaldehyde for different type of vehicles as a function of average speed is presented in Fig. 2 for Euro 2 technology.

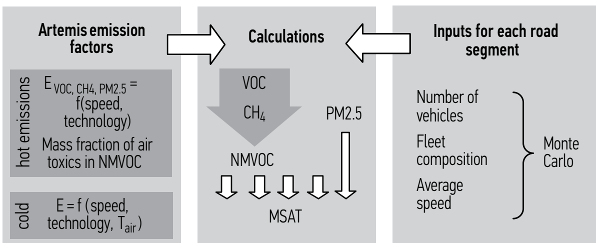

Fig. 1. Calculation algorithm for hazardous air pollutants implemented in TREM–HAP model

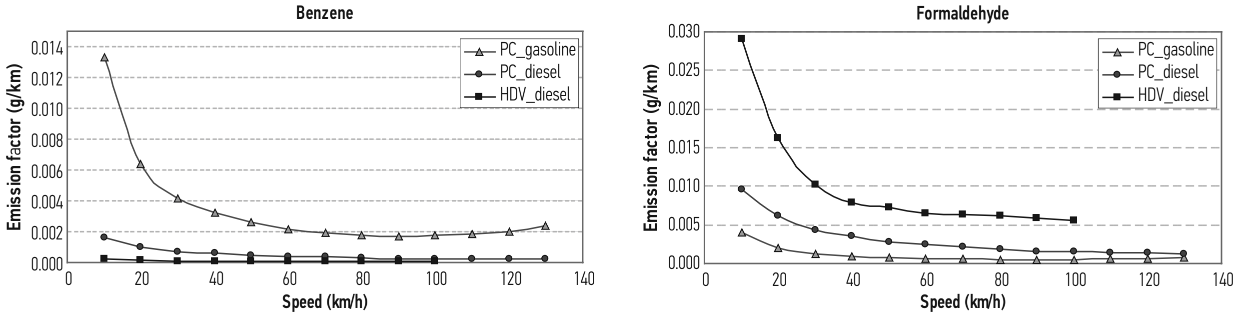

Fig. 2. An example of emission factors for benzene and formaldehyde considered by the emission model for Euro 2 vehicles (PC_gasoline – passenger gasoline cars; PC_diesel – passenger diesel cars with engine capacity < 2 ltr; HDV_diesel – heavy duty diesel vehicles < = 7.5 t)

2.2. Hot Emissions

The hot emission of the pollutant $p(E_p$ [g]) for each road segment is estimated by the model as following:

$$E_p = \sum_i \left(e_{ip}(v) \cdot N_i \right) \cdot L,$$

where: $e_{ip}(v)$ is the emission factor [g·km^{-1}] for pollutant p and vehicle class i defined as a function of average speed v [km·h^{-1}]; N_i is the number of vehicles of class i and L is the road segment length [km].

The emission factors depend on average speed, fuel type, engine capacity and emission reduction technology. However, these data are not available for each counting point and statistical information is usually used to characterise vehicle fleet composition. In this context, uncertainty estimation of the resulting emissions became an important issue.

2.3. Cold-Start Emissions

Cold-start emissions are emitted by vehicles under cold engine and are estimated as an excess to the stabilised hot emission levels. The cold-start excess emission is defined as a difference between the total amount of the pollutant emitted between the start time (t = 0) and time t_{cold}, and the amount of pollutant which would be emitted by the vehicle at its normal running temperature during the same time period. Travel distance, average speed and ambient temperature are considered to quantify cold-start emissions for different vehicle technologies. At urban scale, travel distance is often less than the distance necessary to warm up the engine. Therefore, cold emissions are playing a very important role and their contribution to the total emissions could not be neglected.

In this work, the methodology developed by ARTEMIS (André, Joumard 2005) was adapted in order to be compatible with the model conception. For this purpose, original emission factors represented as absolute emissions [g] per cold cycle were transformed to average cold emission factors [g·km^{-1}] within cold distance.

Cold emission factors are calculated as following:

$$e_{cold} = w_{20^{\circ}\mathrm{C},20\ km/h} \cdot f(T,V) \cdot h(\delta) \cdot g(t),$$

where: e_{cold} – excess emission with a cold engine for a trip [g]; V – average speed during cold engine regime [km·h^{-1}]; T – ambient temperature [°C]; $h(\delta)$ – *distance correction factor = distance travelled* (d) / *cold distance* (d_{cold}) [dimensionless]; $w_{20^{\circ}\mathrm{C},20\ \mathrm{km/h}}$ – excess emissions at reference conditions for T = 20°C and V = 20 km·h^{-1} [g]; $f(T,V)$ – correction factor for speed (V) and temperature (T) effects; $g(t)$ – correction factor for the parking time t.

The ARTEMIS methodology to calculate cold distance was used in order to determine the distance necessary to warm up the engine and to stabilise emissions. A schematic representation of the effect of trip length on the emissions for different classes of passenger cars is presented in Fig. 3. As could be seen in the Fig. 3, the emissions will stabilize within the first 5÷10 km after the start that is considered as a 'cold distance'.

The ARTEMIS methodology used to calculate cold-start emissions is available for passenger cars only, because of insufficient data for other categories, and for typical urban driving, which imply that only urban roads were considered (see Section 3.2). The input parameters considered in the determination of cold-start emission factor are presented in Table 1 considering different passenger car emission classes and fuel type. Calculation of the cold-start emission factor is dependent to the ambient temperature and average speed. The calculation algorithm for acetaldehyde, acrolein and formaldehyde is not sensitive to the ambient temperature. In addition, it should be noted that 1,3-butadiene emissions are totally attributed to gasoline vehicles, while PM2.5 is mainly related with diesel engines.

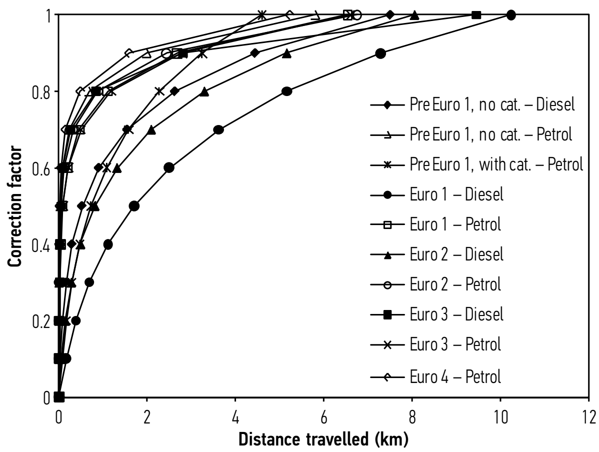

Fig. 3. Schematic representation of the effect of trip length on the cold start excess emissions from passenger cars in winter season

Table 1. Parameters considered for cold-start and hot emission factor quantification

Pollutant	Passenger cars									
	Pre Euro 1		Euro 1		Euro 2		Euro 3		Euro 4	
	gasoline	diesel	gasoline	diesel	gasoline	diesel	gasoline	diesel	gasoline	diesel
PM2.5	–	*T*	–	*T*	–	*T*	–	*T*	–	*T*
Acetaldehyde	V	const.	*V*	const.	V	V	V	V	V	V
Acrolein	–	const.	–	const.	const.	const.	const.	const.	const.	const.
Benzene	*V*, *T*	const.	const.	const.	*V*, *T*	*V*, *T*	*V*, *T*	*V*	*T*	*V*
1,3-butadiene	*V*	–	const.	–	*V*	–	*V*,*T*	–	*T*	–
Formaldehyde	*V*	const.	*V*	const.	*V*	*V*	*V*	*V*	*V*	*V*

Notes: *T*: Ambient temperature (°C); *V*: Average speed ($km \cdot h^{-1}$); const.: constant value; – : methodology is not available

In case of naphthalene, the methodology applied is different and is not presented in Table 1 since the hot and cold emissions are calculated simultaneously and cannot be distinguished.

2.4. Monte Carlo Approach

The Monte Carlo (MC) approach is used to analyse uncertainty propagation, where the goal is to determine how variations in input data affect the emission estimations. For this purpose, a probability distribution is specified for each model input based upon statistical analysis of data. At next, random values are generated for each input parameter taking into account their probability distribution and assuming that the generated values represent real world events. Multiple runs of the emission model based on stochastic inputs provide multiple outputs that can be treated statistically as if they were an experimentally or empirically observed set of data, instead of obtaining a single number for model outputs as in a deterministic simulation (Frey, Bammi 2002).

In the present work, the emissions model has been adapted to use multiple set of randomly generated values for each of the input parameters that characterise the transport activity. Thus, random samples of the number of vehicles, average speed and fleet composition are generated from the respective Probabilistic Density Functions (PDF) and one random value for each input is entered into the model to arrive at one estimate of the model output. This process is repeated over more than 600 iterations to arrive at multiple estimates of the model. These estimates are sample values of the PDF of the model output that reflects the uncertainty in the model inputs.

3. Application

3.1. Study Area

The Porto Urban Area was selected in this study to quantify road traffic emissions of hazardous air pollutants. It is the second largest city in Portugal with a total area of approximately 41 km^2. The resident population of this urban area in 2008 is about 216000 inhabitants (2% of the national population). One of the relevant characteristics of the study area is the centralisation of working places in Porto city centre and an expansion of the agglomeration around the city showing the importance of the population home/work daily trips and consequent air pollution problems in the Region (Tchepel, Borrego 2010).

To study atmospheric emissions induced by transport, the road network was subdivided into 3 types: urban streets, interurban roads and highways with the total length of 78.3 km, 29.8 km and 22.3 km respectively (Fig. 4a). As a total, 84 points distributed within the domain were considered to characterize traffic volume fluctuations. For this purpose, traffic data collected

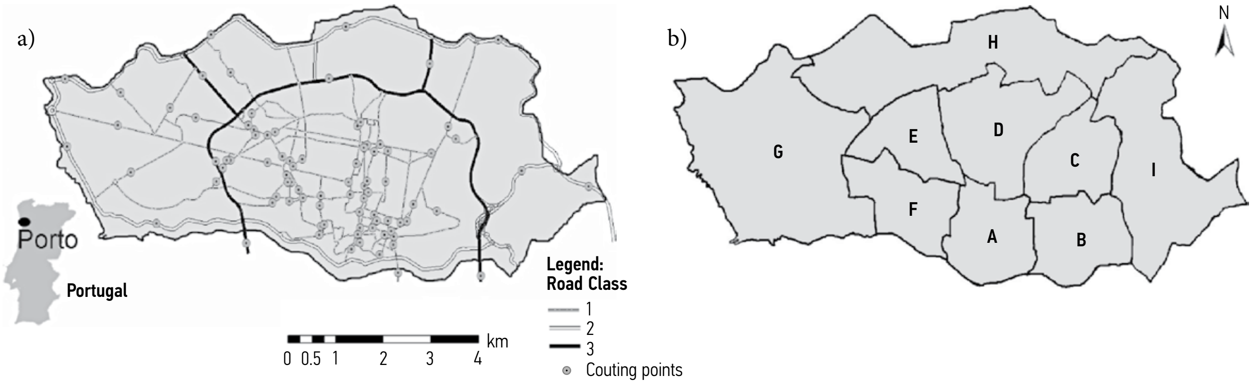

Fig. 4. Administrative limits of the Porto Urban Area and road network considered in the study (type 1 – urban streets, type 2 – interurban roads, type 3 – highways) (a); sectors limits considered in the O/D matrix (b)

Table 2. Origin/Destiny Matrix for each sector (number of displacements in individual transport) for the morning traffic peak period (7:30÷9:30 h) (Oliveira *et al.* 2007)

OD Matrix	A	B	C	D	E	F	G	H	I	Ext. South	Ext. North	Total
A	269	461	430	1070	565	445	500	523	265	447	1819	6794
B	315	84	357	398	200	108	98	275	168	163	504	2670
C	569	436	304	587	344	299	379	622	265	248	587	4640
D	879	335	676	869	609	653	758	902	198	419	1498	7796
E	603	136	391	526	329	532	730	291	103	106	512	4259
F	500	159	198	431	302	215	779	281	47	170	499	3581
G	1344	300	353	774	859	1255	406	1298	135	663	2527	9914
H	855	445	795	1053	639	672	652	582	325	456	1603	8077
I	371	396	383	416	208	204	138	265	100	81	319	2881
Ext. South	1686	998	810	1542	1093	1427	906	735	382	8	14400	23987
Ext. North	7168	2198	3280	3737	2166	4127	4493	4549	1208	11455	11021	55402
Total	14559	5948	7977	11403	7314	9937	9839	10323	3196	14216	35289	130001

by automatic measurements during winter (January and February) and summer (July and August) periods of 2008 were attributed to the road links using road classification and the proximity criteria.

Additionally, population mobility data concerning Origin/Destination trips for traffic peak hours (Oliveira *et al.* 2007) was considered for the study area and subdivided in 9 sectors (Fig. 4b, Table 2). These statistical data provide important information for quantification of cold start emissions as described in Section 3.2.

3.2. Input Data

In order to characterize the uncertainty in input parameters, a set of random inputs characterizing the fleet composition, traffic flow and vehicles speed are generated for each road. The PDF for vehicle classes is determined using the statistical information on vehicle registers and average number of kilometres travelled. For the traffic volume, data from the counting points attributed to each link were used, describing both temporal and spatial variations (Fig. 5). Due to absence of vehicles speed measurements, this variable is estimated for each road segment considering the type of the road and taking into account the speed traffic behaviour adapted from

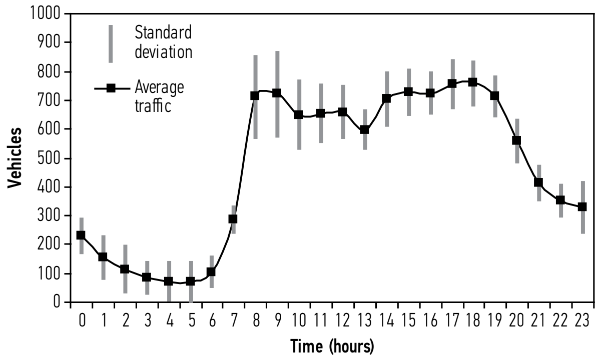

Fig. 5. An example of temporal variation of the passenger car flows obtained from the automatic counting data at a fixed point

Joumard *et al.* (2007): urban (30±9.4 km·h^{-1}), interurban (70±17.6 km·h^{-1}) and highways (110±8.8 km·h^{-1}). A combination of random values generated by the Monte Carlo approach is used to create 625 independent inputs for each road segment to be used by TREM–HAP for the emission estimations.

To estimate excess cold start emissions, a number of vehicles with cold engine have to be considered for each urban road segment. However, it is not possible to obtain this information directly from the automatic traffic counts that is why additional information is required. For this purpose, the ARTEMIS methodology (André, Joumard 2005) to calculate cold distance was used in order to determine the distance necessary to warm up the engine and to achieve a constant emission level (Fig. 3). The statistical information on Origin–Destination (O–D) mobility (Oliveira *et al.* 2007) was considered to determine the daily number of cold starts and the distance between the origin and destination points. Stop duration of 7 hours between the morning and evening peak hours was assumed to calculate the correction factor for cold start emissions. Based on this information, the number of *vehicle* × km performed with a cold engine and a proportion of cold/hot driving was calculated for each urban zone and attributed to the road network.

4. Results

The probabilistic emission inventory for the mobile source hazardous air pollutants was developed based on probabilistic activity factors. It should be stressed that uncertainty of the emission factors was not considered in the current simulations due to absence of the information. Therefore, the overall uncertainty of the emissions is related to the uncertainty in activity data only. The analysis of results examines the influence of the seasonal variations (summer and winter periods), the contribution of hot/cold start to the total daily emissions, the differences of road types and the spatial distribution of the total emissions over the study domain.

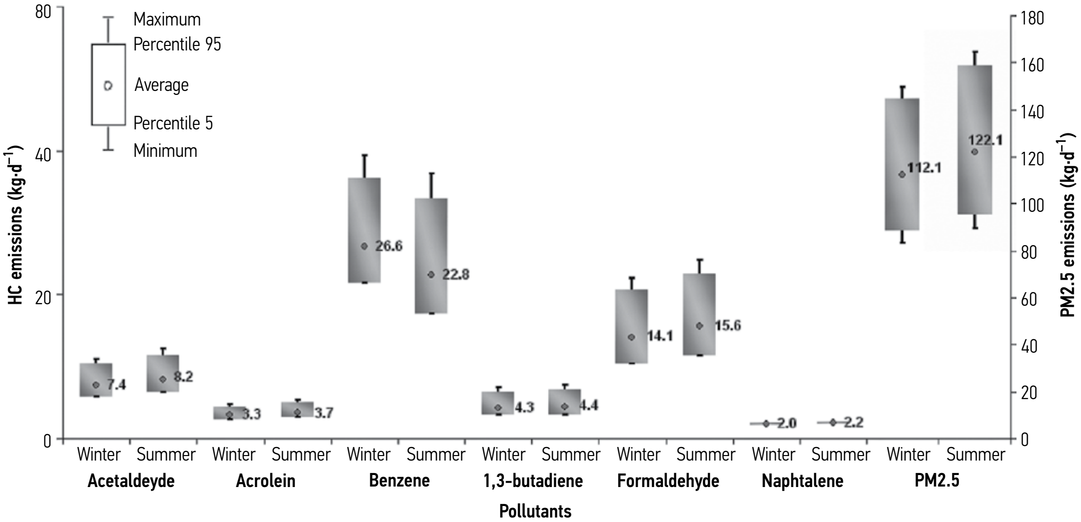

Fig. 6. Statistical parameters for total daily emissions in the Porto Urban Area considering winter and summer periods

The absolute values for total daily emissions estimated for the Porto Urban Area are presented in Fig. 6. Several statistical parameters, including average emissions, 5th and 95th percentile and extreme values were analysed for the selected hazardous pollutants. Also, seasonal difference between summer and winter are examined. It is apparent that PM2.5 and benzene have the largest absolute uncertainty in the daily emissions. For all the pollutants, except benzene, the absolute values for total daily emissions are larger in summer. Benzene has a different seasonal behaviour because of the important contribution of cold start emissions as observed in Fig. 7.

The 90% probability range of the emission estimates are given in Table 3 considering different types of roads. For all the pollutants, urban streets are characterised by higher uncertainty in the emissions achieving the largest range for 1,3-butadiene (–33% to +70 %), while estimations for highways are more robust. Benzene emissions from urban roads are less uncertain than other hydrocarbons, except naphthalene, due to the important proportion of cold start emissions with lower sensitivity to the input data. The very low uncertainties obtained for naphthalene are explained by the different methodology applied for this pollutant. The hot and cold emissions are calculated simultaneously and cannot be distinguished. Also, the methodology to calculate naphthalene emissions is not sensitive to ambient temperature and speed.

The contribution of cold emissions to the total emissions estimated in the study area at typical summer and winter days is presented in Fig. 7.

The results show that the contribution of cold start emissions to the total values calculated for the urban area can achieve 45% in case of benzene, while for other hazardous pollutants this contribution is below of 10% with the only exception of 1,3-butadiene. In general, excess cold start emissions from diesel vehicles are less significant compared with those from gasoline vehicles. As expected, the cold emissions are higher in winter than in summer season due to the direct influence of ambient temperature. However, in the case of acetaldehyde, acrolein and formaldehyde this difference is related to traffic fluctuations only because the calculation algorithm for these pollutants is not sensitive to the ambient temperature. It should be noted that 1,3-butadiene emissions are totally attributed to gasoline vehicles, while PM2.5 is mainly related to diesel engines.

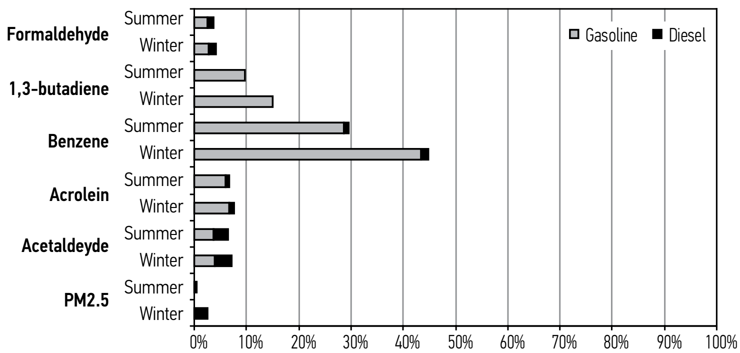

Fig. 7. Contribution of the cold start emission (average values, percentage) to the total emissions within the modelling domain

Table 3. Results of the uncertainties in the emission rates (hot+cold) for the different types of roads

Pollutant	90% probability range of the emission estimates [%]*					
	Urban streets		Interurban roads		Highways	
	(–)	(+)	(–)	(+)	(–)	(+)
PM	–28.1	44.7	–11.6	30.9	–15.7	8.8
Acetaldehyde	–24.7	50.7	–16.3	28.1	–2.0	1.7
Acrolein	–26.6	53.2	–14.9	25.3	–2.0	1.7
Benzene	–22.6	43.4	–23.7	40.2	–5.3	6.1
1,3-butadiene	–33.2	70.4	–21.3	36.5	–3.0	3.5
Formaldehyde	–36.8	65.7	–16.7	28.6	–2.1	2.0
Naphtalene	–0.7	0.6	–0.7	0.6	–0.8	0.7

Notes: (–) = ((5th percentile–mean) / mean) × 100; (+) = ((95th percentile–mean) / mean) × 100

Additionally, the spatial distribution of the daily emissions (hot + cold) was analysed for the study area. Examples for benzene and PM2.5 are presented in Fig. 8.

A different spatial pattern is observed for these two pollutants. Within the Porto Urban Area the highest emission rates of PM2.5 are estimated for highways due to intense traffic during the day.

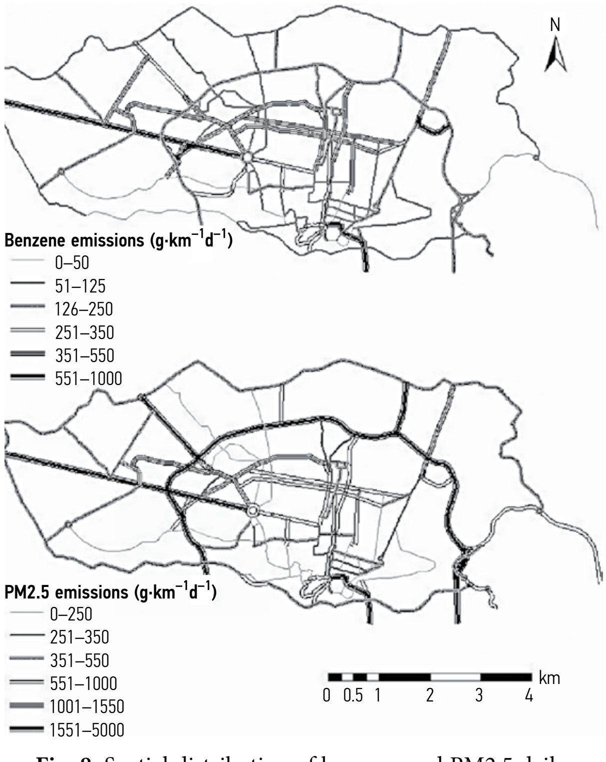

Fig. 8. Spatial distribution of benzene and PM2.5 daily emissions (average) in the modelling domain

Oppositely, benzene emissions are more pronounced at urban streets where the contribution of cold start emissions is very important. For both pollutants, high emissions are obtained in two urban roads which are important thoroughfares connecting the urban centre with peripheral interurban and highway roads.

5. Conclusions

The TREM-HAP model has been developed to estimate the emissions of hazardous air pollutants related to the traffic activity in urban areas. The current work provides a description of the methodology and an application example to characterise a probable distribution of the emissions for different types of roads considering vehicle technology mix, driving conditions and traffic volume fluctuations.

The total daily emissions of air toxics are presented for the entire study area considering their seasonal variations. Different trend is identified for benzene showing 17% higher emissions at winter time due to important contribution of cold starts while other toxic pollutants are mainly affected by changes in the traffic volume that results in higher emissions during the summer period.

Highly uncertain emission data are obtained for the urban roads with the largest range for 1,3-butadiene (–33% to +70%). Oppositely, emissions calculated for highways are generally characterised by a very small uncertainty (less than ±5%) except for PM2.5 (–16% to +9%).

The study shows that cold start emissions can contribute up to 45% to the total daily emissions, highlighting the importance of accounting for cold start emissions in a traffic-related emissions inventory development.

Globally, the results demonstrated that the range of the uncertainty produced in the model application depends on uncertainties in the model inputs but sensitivity of the modelling approach is different for the considered air toxics.

The modelling tool developed and applied in the present work provides spatial distribution of the air toxic emissions for urban areas with complex road network. This information is essential to be used as an input to air pollution models and further population exposure studies. Finally, quantification of the uncertainty range for the emissions opens a possibility to implement air pollution modelling for the study area using probabilistic approach.

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