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Spatial inter-comparison of Top-down emission inventories in European urban areas

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ABSTRACT

This paper presents an inter-comparison of the main Top-down emission inventories currently used for air quality modelling studies at the European level. The comparison is developed for eleven European cities and compares the distribution of emissions of NO_x, SO_2, VOC and $PPM_{2.5}$ from the road transport, residential combustion and industry sectors. The analysis shows that substantial differences in terms of total emissions, sectorial emission shares and spatial distribution exist between the datasets. The possible reasons in terms of downscaling approaches and choice of spatial proxies are analysed and recommendations are provided for each inventory in order to work towards the harmonisation of spatial downscaling and proxy calibration, in particular for policy purposes. The proposed methodology may be useful for the development of consistent and harmonised European-wide inventories with the aim of reducing the uncertainties in air quality modelling activities.

1. Introduction

Emission inventories represent one of the key datasets required for air quality studies, but they are often recognised as the most uncertain input in the modelling chain (Borge et al., 2014; Guevara et al., 2013; Thunis et al., 2016a; Viaene et al., 2013) as their accuracy greatly varies with the type of pollutant, the activity and the level of spatial disaggregation (Davison et al., 2011). In Europe, this is largely due to the fact that regional and local emission inventories are managed and compiled by several different agencies which rely on different standards, methods and categories. This may be understandable given the different background and scope of the inventories, however it may yield to a heterogeneous and inconsistent picture when collating these data for use in modelling at a larger scale (continental and national levels). Furthermore, it is known that, in emission inventories, different measurement methods are applied for the same sectors, e.g. residential combustion which may result in emissions different up to a factor 5 (Denier van der Gon et al., 2015).

For this reason, there exist several top-down implementations that compile EU wide inventories by downscaling national emissions data at a finer resolution: EDGAR (Crippa et al., 2016; Janssens-Maenhout et al., 2017), HTAP_v2 (Janssens-Maenhout et al., 2015), TNO-MACCII and MACCIII (Kuenen et al., 2014, 2015), E-PRTR (Theloke et al., 2009, 2012), JRC07 (Trombetti et al., 2017). These inventories are all comparable in spatial (i.e. between ~10 km x ~10 km and ~7 km x ~7 km) and temporal terms (i.e. annual), geographical extent (i.e. European continent) and thematic resolution (sectors and macro-sectors aggregation) but differences remain in terms of national total emission estimates and/or spatial gridding methodologies. The first type of difference can be caused by model settings, reporting of emission sources, gap filling approaches, assumptions or arbitrary choices and has already been discussed for some inventories (Kuenen et al., 2014; Granier et al., 2011).

For the second difference, spatial discrepancies mostly depend on methodological assumptions, proxies' availability and choice of the weighting methodology. The fact that all these inventories are developed at a high spatial resolution (~7–10 km x ~7–10 km) reinforces this factor. As shown by Zheng et al. (2017), the spatial mismatch between gridded inventories developed from different spatial proxies is largely diminished at coarse resolutions (i.e. 25 km × 25 km) but

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tends to increase as grid size decreases (i.e. 4 km × 4 km).

These differences have often been overlooked and only studied for regional (i.e. sub-national) inventories (Winiwarter et al., 2003; Vedrenne et al., 2016) while only a few cases at fine scale have been published (Ferreira et al., 2013). These studies clearly stressed the importance of the assumptions behind the underlying proxies, their level of detail and their accuracy, to explain the very low spatial correlations found between target inventories. It is important to note that these spatial variations have a strong impact on air quality modelling results (Geng et al., 2017; Zhou et al., 2017), especially when the results are considered for policy making and planning options. Top-down emission inventories are often being used as input data for modelling activities at urban scale (López-Aparicio et al., 2017); therefore, particular attention should be given before choosing a specific dataset for this kind of modelling activities.

To our knowledge, our study is the only existing spatial intercomparison between emissions inventories currently used at the European scale. Its novelty lies on defining the possible uncertainties in the spatial proxies behind the disaggregation and allocations of emissions in urban areas and, consequently, on reducing the propagation of errors to air quality models and their applications.

This study assesses how a set of six EU wide emission inventories (i.e. EDGAR, TNO_MACCII, TNO_MACCIII, INERISinv, EMEP, JRC07) behave in selected European urban areas in terms of sectorial shares and regional allocation also through the application of a novel approach, namely the diamond analysis (Thunis et al., 2016b), in order to estimate systematically the spatial variability between them. This approach aims to contribute to increasing the reliability of emission inventories. We first describe the methodology and the emission datasets used, before identifying the main differences for the selected urban areas. Finally, recommendations to improve credibility for air quality modelling applications and reduce the level of uncertainty are provided for each inventory.

2. Methodology

We focus our analysis on the way emissions of NO_x, SO_2, VOC and $PPM_{2.5}$ are spatially distributed by different European scale top-down inventories. For this reason, the comparison is not made in terms of absolute, but rather in terms of normalised emission values. The values attributed to each grid cell of coordinates i and j for the variable $E^*_{s,p}$ represent the percentage of the total national emission for each emission pollutant “p” and sector “s”, i.e.:

$$\forall\, s\,,\ \forall\, p\!:\ E^*_{s,p}(i,j) = \frac{E_{s,p}(i,j)}{E^{tot}_{s,p}}$$

where $E^{tot}_{s,p}$ represents the country total emission for a given sector and pollutant. With this normalisation, observed differences between inventories at a given grid cell do not depend on the original national emission value, but instead depend on the downscaling methodology and ancillary data used (Hiller et al., 2014).

The spatial analysis is performed for specific urban areas and for the main emission macro sectors: non-industrial combustion (SNAP02), industrial activities (SNAP03 and SNAP04, which are kept together in order to facilitate the comparison within inventories: SNAP34) and road transport (SNAP07). See the Supplementary Information (SI) for a description of the SNAP Macro Sectors (Table 1, SI).

The SNAP02 macro-sector consists of i) commercial/institutional stationary combustion; ii) residential combustion; iii) stationary combustion associated with agriculture, forestry or fishing; iv) other stationary. Given that the sector “ii) residential combustion” is the dominant one, the discussion in this paper focuses only on this subsector, hereafter referred to as ‘Residential’.

Eleven cities (Barcelona, Bucharest, Budapest, Katowice, London, Madrid, Milano, Paris, Sofia, Utrecht and Warsaw) were selected across Europe to represent the diversity of environmental and anthropogenic factors (i.e. meteorology, economic activities, energy system, population density and land use) over the continental domain; in particular, the differences in Land Use cover reported in Table 2, SI, will affect the sectorial shares of emissions in each study site. For each city, the study area covers approximately 35 × 35 km^2, including only whole grid cells without having to split or resample them. With the exception of EDGAR, all inventories have similar spatial resolution and grid alignment, so it was possible to define common study areas. The EDGAR inventory has a different spatial resolution and so an alternative definition of the study areas was created resembling the original one, while preserving the integrity of the selected grid pixels. The standard study site and the adjusted EDGAR one for each urban area are shown in the SI with the considered land use pattern (Figs. 1 and SI).

The assessment is supported by the analysis performed by means of the diamond approach (Thunis et al., 2016b), a novel method which, by using total emission ratios, allows the comparison of emission inventories and the identification of the likely cause (activity level or activity share) of differences between them. Given the normalisation by the country totals, the differences seen among inventories in terms of activity levels and share can be directly attributed to the spatial disaggregation methodology.

2.1. Downscaled inventories

We consider six European scale top-down inventories, with 2010 as reference year, unless mentioned otherwise. The selected emission inventories cover a wide and important range of applications, including regulatory purposes (e.g. EMEP), monitoring services (e.g. TNO-MACC, EDGAR) and integrated assessment (e.g. INERISinv, JRC07).

- EDGAR version v4.3.1, January 2016 (European Commission, 2016a; Crippa et al., 2016), hereafter referred to as EDGAR. This inventory provides global emissions for gaseous and particulate air pollutants (BC, CO, NH_3, NMVOC, NO_x, OC, PPM_{10}, $PPM_{2.5}$, SO_2) per IPCC sector (Intergovernmental Panel on Climate Change) covering the whole time-series 1970–2010 at the global scale. Emissions are provided in tons of substance at 0.1 × 0.1° resolution. A highly detailed re-mapping of the sectors from the IPCC to the SNAP nomenclature has been made to allow comparing with the other databases. The simplified version of the mapping scheme from IPCC to SNAP codes is included in the SI (Table 3) together with the detailed reclassification for a representative SNAP MacroSector (SNAP04, Production Processes, Table 4, SI).
- TNO-MACCII (Denier van der Gon et al., 2010; Kuenen et al., 2011, 2014), hereafter referred to as MACCII. The TNO emission inventory was developed for Europe by TNO for the years 2003–2009. It has a 1/8° longitude x 1/16° latitude resolution and covers NO_x, SO_2, NMVOC, NH_3, CO, PPM_{10}, $PPM_{2.5}$ and CH_4. This dataset is not available for 2010, consequently the 2009 dataset has been used instead.
- TNO-MACCIII (Kuenen et al., 2014, 2015; MACC-III Final Report, 2016), hereafter referred to as MACCIII. It is the updated version of the TNO-MACCII product, which extended the time-series from year 2000 to year 2011. All years were revisited and the spatial distribution proxies updated and improved, often based on user comments.
- INERISinv (hereafter referred to as INERIS): The INERIS inventory is based the work by Bessagnet et al. (2016) with the following changes for the Macro Sectors analysed in this work.
  MS34: The E-PRTR database is used for Large Point Sources of emissions (Mailler et al., 2017)
  MS07: Road transport emissions of all considered countries are distributed using a proxy based on the combination of several databases and the French bottom-up emission inventory (Mailler et al., 2017)

MS02: Residential combustion emissions are distributed based on population, land use and the French bottom-up emission inventory. For all compounds, except PPM, emissions are redistributed according to the population distribution. For $PPM_{2.5}$ the method proposed by Terrenoire et al. (2015) using a logarithmic regression as a function of population density was improved by fitting several parameters to the new proxy and the landuse to treat differently urban areas from non-urban areas.

For all sectors and for all compounds, the French and British bottom-up emission inventory at 1 km resolution is used to redistribute emissions of France and United Kingdom.

The inventory covers NOx, SO_2, NMVOC, NH_3, CO, PPM_{10} and $PPM_{2.5}$ as reported by each country in EMEP. It is distributed at a 1/8° longitude x 1/16° latitude resolution and it covers the year 2010.

- EMEP: The EMEP emission inventory is based on emission data reported by the 51 Parties belonging to the LRTAP convention, complemented by expert estimates (www.emep.int). The EMEP emission product is normally distributed at 50 × 50 km^2 resolution and calculated by using sectoral emissions as reported by countries and gap filled with data from different models where no or incomplete data are reported by countries (EMEP, 2015). At the 36th session of the EMEP Steering Body, the EMEP Centers suggested to increase spatial resolution of reported emissions from 50 × 50 km^2 to 0.1° × 0.1° (http://www.ceip.at). Nevertheless, the official reporting of gridded emissions in this new resolution is requested from 2017 onwards and currently about half of the EU28 countries have submitted their own gridded data. The higher resolution version used here has instead been rescaled at 1/8° longitude x 1/16° latitude resolution based on the TNO-MACCIII emission data and it covers year 2013.
- JRC07 (Trombetti et al., 2017). The JRC07 is an inventory recently developed for use in Integrated Assessment Modelling strategies (IAM) in the fields of regional air-quality (Clappier et al., 2015; Carnevale et al., 2012) and land use and territorial modelling (https://ec.europa.eu/jrc/en/luisa; Lavalle et al., 2011; Lavalle et al., 2013). The inventory is based on country total emission data from the Greenhouse Gas and Air Pollution Interactions and Synergies Model (GAINS, Amann et al., 2011), as used for the implementation of the EU20-20-20 targets under the assumptions of the 2013 Air Quality package review. It models emissions from 2010 up to 2030 and it currently covers NO_x, SO_2, VOC, PPM_{10}, $PPM_{2.5}$ and NH_3. It is distributed with different spatial resolutions and here it is being used in its version at 1/16° x 1/16° resolution.

An overview of the spatial proxies and ancillary data used for the spatial distribution of emissions from the considered sectors is shown in Table 1.

3. Analysis

3.1. Comparison at country scale

The selected inventories are first analysed and compared in terms of input data, looking at their macro-sectorial shares of emissions aggregated at the EU28 level ($PPM_{2.5}$ is represented in Fig. 1; see Fig. 2 in the Supplementary Information for the other pollutants). It is important to underline that TNO-MACCII, TNO-MACCIII, INERIS and EMEP are based on the officially reported emissions by the countries to the CLRTAP, while JRC07 is based on GAINS. Although the reporting year to CLRTAP might not be the same for all inventories, there is a good agreement among the inventories based on official reporting to CLRTAP on the shares of emissions for the targeted macro sectors. The EDGAR emission inventory shows the largest differences for all pollutants with the exception of SO_2.

For NO_x, the share for road transport varies from 37% (EDGAR) to 43% (INERIS), while differences are between 1% and 2% for the residential combustion and the industrial sectors. The most noticeable difference for NO_x is in the EDGAR emission inventory, as it assigns more emissions (~7%) to SNAP01 (Combustion in energy and transformation industries), which is compensated by a lower share of emissions in SNAP08 (Non-Road transport).

For $PPM_{2.5}$, the share of emissions from SNAP02 (Residential) ranges from 38% (EDGAR) to 48% (GAINS, on which JRC07 is based) with the exception of EMEP, which assigns much more importance to this sector (54%). This difference between EMEP and the other CLRTAP-based inventories for $PPM_{2.5}$ can be explained by different emission reporting in different years. The EMEP inventory is based on reporting in 2016 while e.g. TNO-MACCIII is based on reporting in 2013. Overall EU28 reported primary $PPM_{2.5}$ emissions from SNAP02 in 2016 are more than 20% higher than in 2013.

For SNAP07 (Road Transport) and SNAP34 (Industry), we find the same pattern reported for NO_x, with EDGAR assigning to industry 5% higher emissions than MACC2 and ~10% higher than the other inventories, while reporting a ~6% lower share of the road transport sector. Similar variations are also observed for the agricultural sector (SNAP10).

In the case of SO_2, no major difference is observed between the inventories, although it has to be noted that the road transport sector is of negligible importance. Emissions of SO_2 have decreased by 88% between 1990 and 2014 in EU28 mainly as a result of fuel-switching from high-sulphur solid and liquid fuels to low-sulphur fuels (EEA, 2016) but also as a result of the increase in abatement on large plants. Currently, emissions from this pollutant mainly come from point sources linked to the public energy production sector (i.e. coal-fired power plants) that are usually continuously monitored and hence well characterised by all emission inventories. This might not apply though for some other countries where, with the above mentioned increased abatement and fuel switch, the share of emissions of the national total from Large Point Sources has significantly decreased and a higher share of emissions come from Medium Combustion Plants (MCPs) and possibly even from small-scale combustion.

Looking at the target sectors for VOC, while there is a good agreement for SNAP02 and SNAP07, EDGAR has higher emissions for the industrial sector. This is most likely an allocation issue, since this difference is partially compensated by an underestimation in SNAP06 (Solvents and other Products use). This compensation between sectors may indicate a potential inconsistency in the mapping of industrial activities related to the use of solvents (e.g. pharmaceutical products, paint manufacturing). This inconsistency highlights that differences in the original mapping and linking tables used in each inventory to match specific pollutant activities to an official reporting format (e.g. NFR to SNAP) may have a large impact when re-mapping activities from one reporting nomenclature to another.

3.2. Comparison at regional/urban scale

3.2.1. Emission totals

We focus here on the regional allocation of emissions, i.e. on the fraction of the sum of national emissions from SNAP02, SNAP34 and SNAP07 which is assigned to a particular city (Fig. 2; as in the following figures, the cities are ordered on the *x* axis by degree of longitude, West to East). All inventories perform similarly for NO_x with an exception in Budapest to which EDGAR assigns almost 30% of the national totals, almost twice the percentage assigned by the other inventories. Budapest consistently shows the largest differences between the inventories for all compounds. Large differences are also observed for Paris, for all pollutants and especially for EDGAR and MACCII, and in Bucharest and Sofia, for SO_2 and $PPM_{2.5}$. The higher emission share in Paris according to MACCII could be explained by an over-allocation of industrial emissions (SNAP34) to urban areas. Emissions from the industrial sectors that cannot be linked to a specific point source are merged in MACCII and gridded based on total population (Table 1). This approach

Table 1
Description of the features of the inventories (literature reference and temporal coverage), spatial proxies and ancillary data used for the downscaling of emissions from the considered sectors.

	TNO-MACCII	TNO-MACCIII	INERISinv	JRC07	EDGAR	EMEP
Reference	Kuenen et al., 2014	Kuenen et al., 2015, Kuenen et al., Pers. Comm	Mailler et al., 2017	Trombetti et al., 2017	Crippa et al., 2016	Mareckova et al., 2016
Temporal Coverage	2003–2009	2000–2011	2010	2010–2030	1970–2010	2013
Input Emission Data	Official reported emissions to CLRTAP, gapfilled with other data (e.g. GAINS, EDGAR, TNO estimates)	Official reported emissions to CLRTAP, gapfilled with other data (e.g. GAINS, EDGAR, TNO estimates)	EMEP	GAINS	EDGAR	Official reported emissions to CLRTAP, gap-filled by CEIP
SNAP02 (Residential)	Total population (CIESIN and GRUMP http://sedac.ciesin.columbia.edu) and wood use map (based on population and wood availability)	Total population (CIESIN and GRUMP http://sedac.ciesin.columbia.edu) and improved wood use map (based on population and wood availability)	Population (GRUMP http://sedac.ciesin.columbia.edu/gpw) Land Use (USGS)	Population, Industrial and Agriculture Land Use (LUISA, Lavalle et al., 2013) Degree of Urbanization (EUROSTAT, http://ec.europa.eu/eurostat/web/degree-of-urbanisation/overview)	In-house proxy based on rural and urban population (http://sedac.ciesin.columbia.edu/)	50 × 50 km_2: CEIP country reported grids 0.0625 × 0.125°: TNO-MACCIII emission data
SNAP03 and SNAP04 (Industry)	LPS: E-PRTR, TNO point source database Diffuse: Population	LPS: E-PRTR, TNO point source database Diffuse: Industrial land cover (CORINE (EEA, 2017))	LPS: E-PRTR Diffuse: No Diffuse	LPS: E-PRTR v8 Diffuse: Manufacturing sector employment data (Eurostat, 2008), Industrial Land Use (LUISA, Lavalle et al., 2013)	LPS: In-house proxy based on USGS (http://mrdata.usgs.gov/mineral-operations/), E-PRTR v4.2, v6.1, v7 CEC (http://takingstock.cec.org/), CIESIN (http://sedac.ciesin.columbia.edu), Global Energy Observatory (http://globalenergyobservatory.org/), NGDC (https://www.ngdc.noaa.gov/eog/viirs.html; https://www.ngdc.noaa.gov/eog/viirs.html), World Port Index (PUB 150) (http://msi.nga.mil/MSISiteContent/StaticFiles/NAV_PUBS/WPI/Pub150bk.pdf) Diffuse: No Diffuse Population when no LPS is available	50 × 50 km_2: CEIP country reported grids 0.0625 × 0.125°: TNO-MACCIII emission data
SNAP07 (Road Transport)	TRANSTOOLS network (European Commission, 2005) and Total population	TRANSTOOLS network (European Commission, 2005) and Total population	Proxy based on the correlation between the French bottom-up emission inventory and different spatial databases (CORINE, EEA; ETISplus, http://www.etisplus.eu)	Open Street Map Network (OSM contributors, 2015) Population (LUISA, Lavalle et al., 2013) TREMOVE shares of traffic (De Ceuster et al., 2006) AADT UNECE (UNECE, 2005)	In-house EDGAR proxy based on OpenStreetMap (OSM contributors, 2015) and weighted on road type and vehicle category	50 × 50 km_2: CEIP country reported grids 0.0625 × 0.125°: TNO-MACCIII emission data

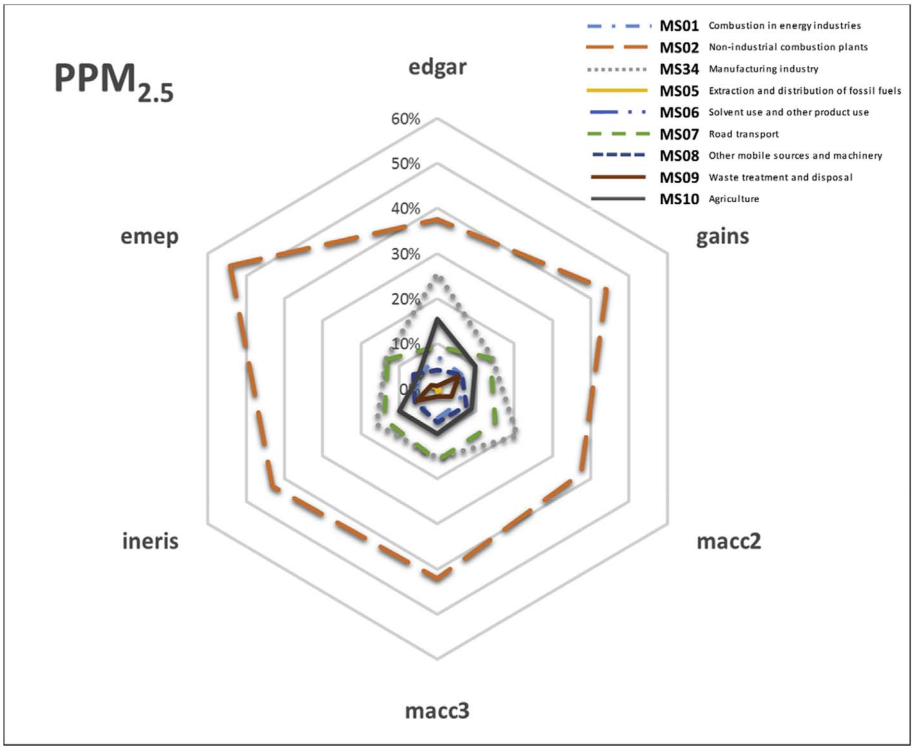

Fig. 1. Comparison of the selected inventories for $PPM_{2.5}$ in terms of macro-sectors shares at the country scale. The numbers from 1 to 10 refer to the SNAP sectors, where SNAP34 is the result of merging SNAP03 and SNAP04.

resulted in an over-allocation of industrial emissions in urban areas (Guevara et al., 2014) and has been corrected in MACCIII where diffuse industrial emissions are allocated to industrial areas according to the CORINE land cover classification 2016 (EEA, 2017b). A good agreement between the inventories is observed in Barcelona, Milano, Warsaw and Utrecht. From an inventory point of view, EDGAR tends to allocate a larger fraction of the national totals to urban areas than the other inventories, in particular for $PPM_{2.5}$ and SO_2. The higher

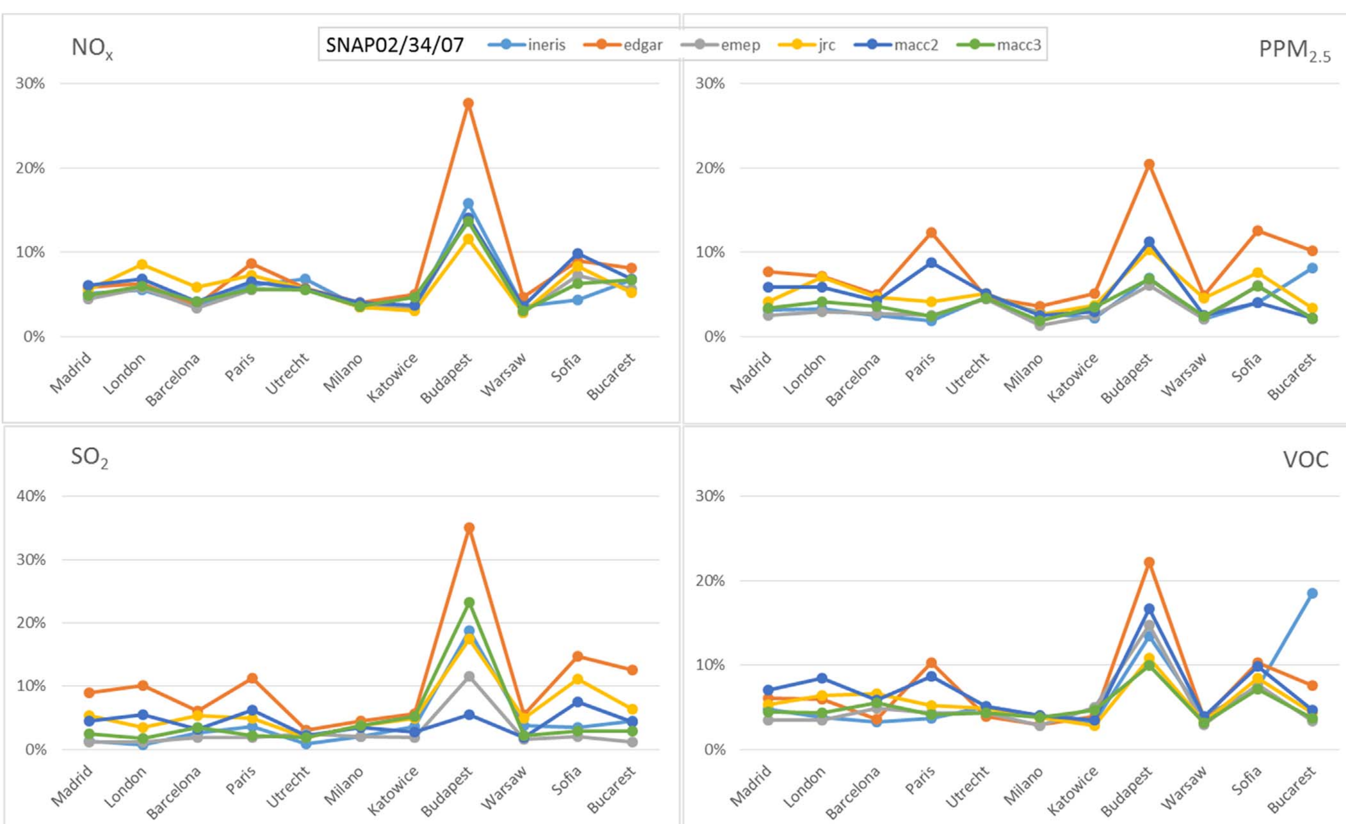

Fig. 2. Regional allocation of emissions: fraction of the country emissions from the total of sectors SNAP02 (Residential Combustion), SNAP34 (Industry) and SNAP07 (Road Transport) which is assigned to each city.

estimation ranges between factors 1.5 and 2. The behaviour of INERIS in Bucharest NO_x and SO_2 follows the average trend while, for VOC and $PPM_{2.5}$, it is outlying. As it appears from the analysis at sectorial level in the next chapters, these higher values are likely due to higher emissions from the industrial sector which represents the ~70% of the total emissions, a share much higher than the ones reported for the other inventories (~5%–~40%).

In general, it is clear that the spatial disaggregation methods applied in each inventory work differently in terms of urban areas and pollutants.

3.2.2. Sector share

In order to better understand why the spatial allocation differs between inventories, we compare the way each inventory spatially allocates the regional emission in terms of macro-sectors, more specifically, transport (SNAP07), industry (SNAP34) and residential combustion (SNAP02). Some uncertainties could be present due to the way different countries might convert sectors between the different nomenclatures (NFR, SNAP, IPCC).

For each urban area, the contribution of each macro-sector, which will partly depend of the characteristics of the selected study site (Table 2, SI), it is assessed in terms of percentage of the total city emission. The regional/city macro-sector percentages (C) are computed as

$$C_m^p = \frac{E_m^p}{\sum_{i=1,M} E_i^p}$$

where E_m^p represents the total city emission for a pollutant "p" and macro-sector "m" and M is the total number of sectors (3 in our case).

In general, NO_x and SO_2 show the most robust trend among the four pollutants, while it is not possible to identify a consistent pattern for VOC in terms of cities or in terms of sectors.

Among the three macro-sectors, the industrial one is by far the least consistent with large differences in many cities (values up to 5 times bigger, Fig. 3). Similar inconsistency was highlighted when comparing regional downscaled inventories with bottom-up emission inventories for the same urban areas (López-Aparicio et al., 2017). While the INERIS inventory has systematically lower values for SO_2, EDGAR tends to allocate higher industrial emissions to most cities for most of the emission pollutants. It is also noticeable that, as noted in the previous paragraph, TNO-MACCIII has reduced the amount of industrial emissions located in urban areas with respect to TNO-MACCII. This also results into a larger relative contribution from SNAP07 in TNO-MACCIII when compared to the previous version.

The transport sector shows the most similar shares across inventories, with the exception of VOC (Fig. 4; SO_2 not shown due to its low importance for this sector). With the exception of two cities (i.e. Sofia for NO_x and VOC and Utrecht for $PPM_{2.5}$), EDGAR systematically allocates a much lower fraction of transport emissions to urban areas. This is probably due to the fact that emissions from on-road transport sector are distributed in EDGAR based on road types and vehicle categories and not considering the population density which is in some way taken into account in the other inventories.

The residential sector (SNAP02) shows good agreement among the inventories for NO_x and, in particular, there is no difference between TNO-MACCII and TNO-MACCIII (Fig. 5). In the case of $PPM_{2.5}$, although the trends are quite consistent, there are differences in terms of percentages, indicating greater variability in emitting sources (Fuelwood, Coal), which are distributed differently by each inventory (Table 1). This is especially true for Eastern European cities, such as Bucharest, Katowice and Warsaw.

The EDGAR emission inventory shows different patterns from all other inventories; in particular, there are larger emission estimates from the residential sector for NO_x and, to a less degree, for some cities for $PPM_{2.5}$ and VOC, which are partially compensated by lower contributions from the road transport emissions.

Fig. 3. Sectorial allocation. Share of the total emissions for each city coming from the Industry sector.

Fig. 4. Sectorial allocation. Share of the total emissions for each city coming from the SNAP07, road transport sector.

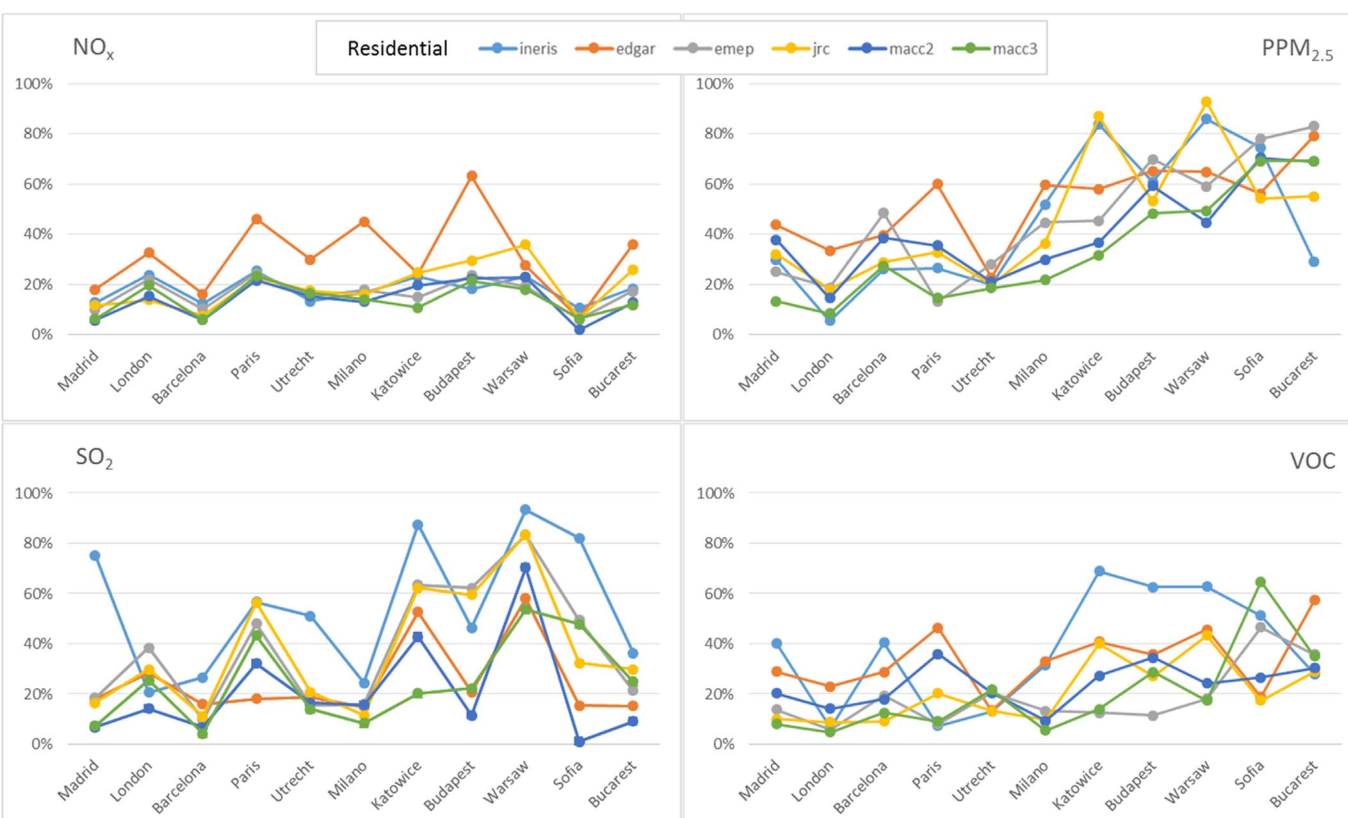

Fig. 5. Sectorial allocation. Share of the total emissions for each city coming from the SNAP02, residential combustion sector.

3.3. Activity/share analysis: the diamond approach

3.3.1. Methodology

Thunis et al. (2016b) proposed a methodology to compare emission inventories for different pollutants (i.e. PPM, NO_x, VOC, SO_2, etc …) and activity macro-sectors (i.e. transport, industry, residential, etc …), on the basis of emission ratios between two inventories. In a first step, the emission for a pollutant p and a macro-sector s ($E_{s,p}$) is expressed as the product of an emission factor (e) and an activity (A). The emission ratio between two inventories then equals to the product of an emission factor ratio and an activity ratio:

$$\hat{E}_{s,p} = \hat{e}_{s,p}\hat{A}_s$$

with $\hat{E}_{s,p} = \hat{E}_{s,p}^{(1)}/\hat{E}_{s,p}^{(2)}$; $\hat{e}_{s,p} = \hat{e}_{s,p}^{(1)}/\hat{e}_{s,p}^{(2)}$ and $\hat{A}_s = \hat{A}_s^{(1)}/\hat{A}_s^{(2)}$ in which superscripts 1 and 2 identify the two inventories for a pollutant p and a macro-sector s.

The methodology detailed in Thunis et al. (2016b) aims to quantify inconsistencies in terms of emission factors and activity ratios ($\hat{e}_{s,p}$ and $\hat{A}_s$) from the limited knowledge we have of the total emission ratio ($\hat{E}_{s,p}$). It assumes that one pollutant species (denoted as p^*) can be identified as reference for which the emission factors are equal in the two inventories (i.e. $\hat{e}_{s,p*} \approx 1$). With this condition, it is then possible to deduce the emission factors and activity ratios from the total emission ratios: $\hat{A}_s \approx \hat{E}_{s,p*}$ and $\hat{e}_{s,p} \approx \hat{E}_{s,p}/\hat{E}_{s,p*}$.

The need to select a reference pollutant is a disadvantage of this methodology as discussed in Thunis et al. (2016b). However, in this work we follow an alternative approach that does not require a reference pollutant. We assume that the activity and emission factor ratios behave as random variables with probability distributions following a Gaussian law centered around 1. These distributions are then used to estimate the probability that the activity and emission factors ratios take specific values within given intervals, while satisfying the known constraint on total emission ratios. The activity and emission factor ratio are then those characterised by the highest probability. The activity and emission factor ratio are used as X and Y coordinates in the "diamond" diagram, where each sector-pollutant couple is represented by a specific point (Fig. 6). As a result of the construction, the diagonals (slope $= -1$) provide information on the overall under-/over-prediction in terms of total emissions. We can define a diamond shaped area where activity, activity shares and total emissions all remain within given degrees of variation. For example, the red diamond indicates ratios of activity, emission factor and total emissions all within 100% (or a factor 2) differences, while the green diamond indicates ratios within 50% (or a factor 1.5). Colors and symbols are used to identify pollutants and sectors, respectively. These choices are made to facilitate the identification of the different ratios. The size of the symbol is then made proportional to the emission magnitude (i.e. the emission for one sector is compared to the total emitted for one given pollutant). This feature helps identify the biggest contributors and potential sectors to mitigate. It has to be in fact remarked that this kind of analysis does not aim to draw final conclusions but is instead a screening tool to highlight possible sources of inconsistencies between inventories. The reader is referred to Thunis et al. (2016b) for more details.

This approach allows us to compare the 6 inventories for 4 pollutants and for 11 cities. However, the "diamond" approach only allows relative comparisons because no emission inventory can be considered as the reference inventory. A synthetic inventory was therefore created for the relative comparison and to be used as a reference dataset. The synthetic emission values are computed as the median values of the 6 existing inventories. The results discussed in the next sections are based on the differences and similarities between the six top-down inventories when compared to the synthetic dataset in terms of emission sector share and activity data (i.e. the "data on the magnitude of human activity resulting in emissions or removals taking place during a given period of time", IPCC, 2006). Even if the median values could be affected by outlying values, the general trends describing the nature of discrepancy between inventories are expected to be anyway meaningful.

It is noteworthy to remark that, in this work, urban emission totals are further scaled by their country totals as explained in the methodology. This step is made to ensure that all urban inventories originate from similar country totals and that the observed differences in the diamond approach focus on the differences in terms of spatial allocation of the emissions rather than on country scale biases. In this particular case, the value on the X axis is now an indication of the differences in terms of activity shares rather than in terms of emission factors.

3.3.2. Analysis in terms of sector

Transport Sector – There is an overall agreement between the inventories both in terms of activity intensity and sectorial share as indicated by the fact that most points are concentrated within the diamond shape (Fig. 6). This is probably explained by the fact that similar proxies are used for the spatial and sectorial disaggregation from the country totals, allowing to allocate similar amounts of emissions to the considered study areas. Indeed, the spatial information related to the road network (e.g. Open Street Map) is one of the most precise and shared pieces of information (especially at the spatial resolution considered in this work). The proxies used to allocate traffic intensity in each inventory are also quite similar and do not impact the emission distribution significantly. Activity level is however lower according to EDGAR (especially Paris, Barcelona) and INERIS (for the Eastern European cities). This pattern is especially visible for VOC, probably related to the way the inventories deal with the evaporative emissions. It is also interesting to note that the diagram does not show the same consistency if city totals are not scaled to the national totals (not shown), indicating that most of the differences between inventories tend to originate from differences in country total estimates rather than from the spatial disaggregation proxies.

Residential Sector – As noted above, the most consistent trends in this sector across cities appear for NO_x, with the exception of EDGAR (Fig. 7) and, for a few cities, of EMEP and MACCIII. The larger differences observed for PPM2.5 and VOC are mostly due to a problem in terms of activity share (points spread along the horizontal axis) rather than in terms of activity intensity. Given the fact that NO_x emissions are more consistent than the other pollutants, the difference must be due to activities which are not a significant source of NO_x emissions, such as wood burning. It is interesting also to note the INERIS behaviour; for most cities in this inventory, the points representative of $PPM_{2.5}$ and VOC are aligned on the same horizontal line, indicating a similar proportional overestimation of the activity (wood and coal burning) in all cities. This similar overestimation probably results from using a parameter proportional to population to scale up wood-burning emissions. In INERIS, emissions from SNAP02 are in fact distributed according to a proxy based on population, land-use and the French bottom-up emission inventory, fitting other parameters in order to differentiate urban and non-urban areas. Differences across the cities are hence mainly due to inconsistencies in terms of shares of activities within the same sector, with a proportion that depends on the importance of wood and coal burning in each city: the higher the importance of wood and coal burning, the higher the uncertainty of emission distribution in this sector. For instance, in countries such as Germany and Spain, emissions from residential heating are lowest, whereas Romania, Poland and France have the highest levels (Terrenoire et al., 2015). This confirms the importance of updating the emission estimates from the residential combustion sector, as stated by Denier van der Gon et al. (2015) and developing a proxy which would allow for a better and common representation of the spatial distribution of wood and coal usage, also taking into account site-specific features such as the proliferation of district heating in many cities which results in a smaller and secondary usage of conventional wood-fired stoves.

Residential Sector: INERIS as a reference inventory - INERIS can be

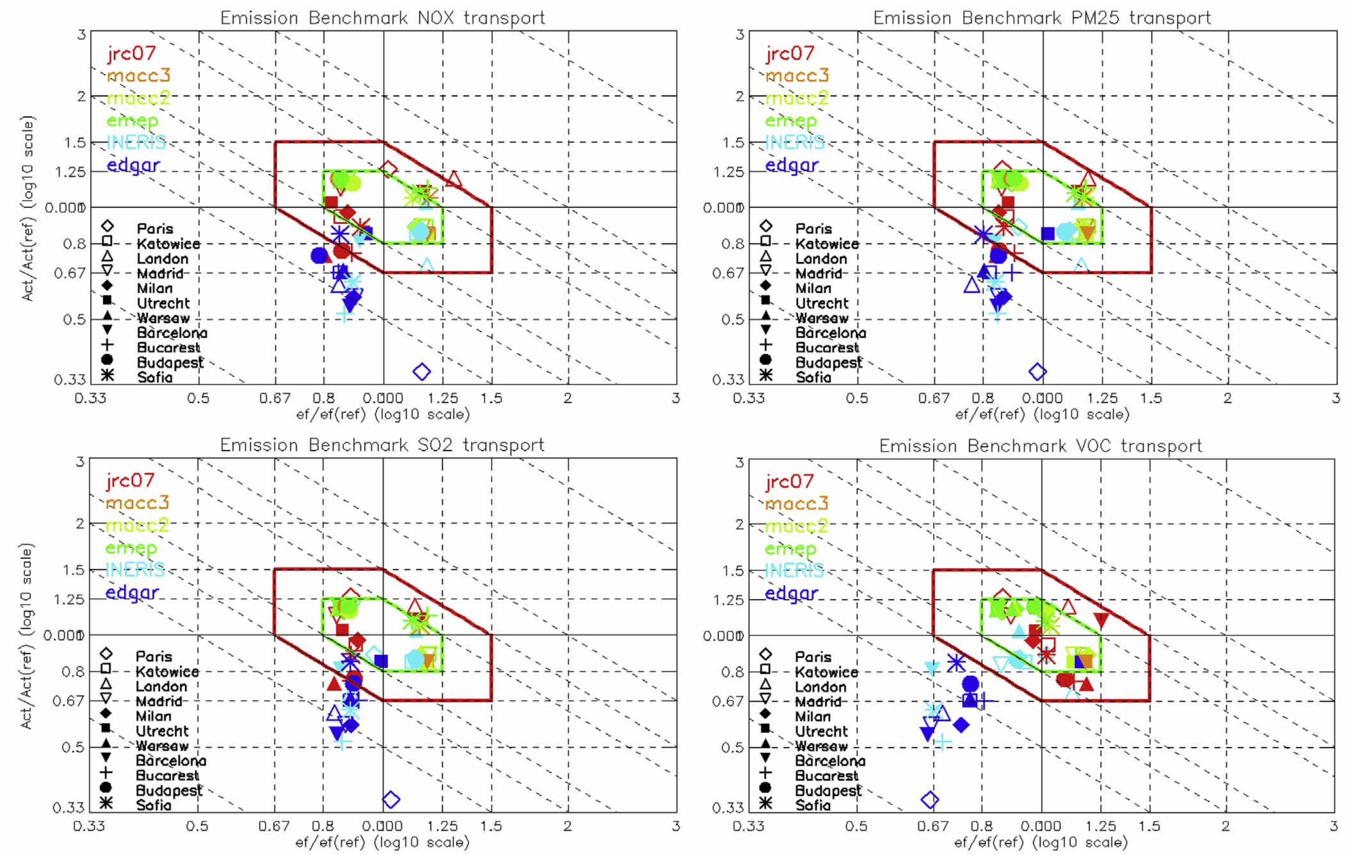

Fig. 6. Comparison between inventories using the diamond approach for the Road Transport sector.

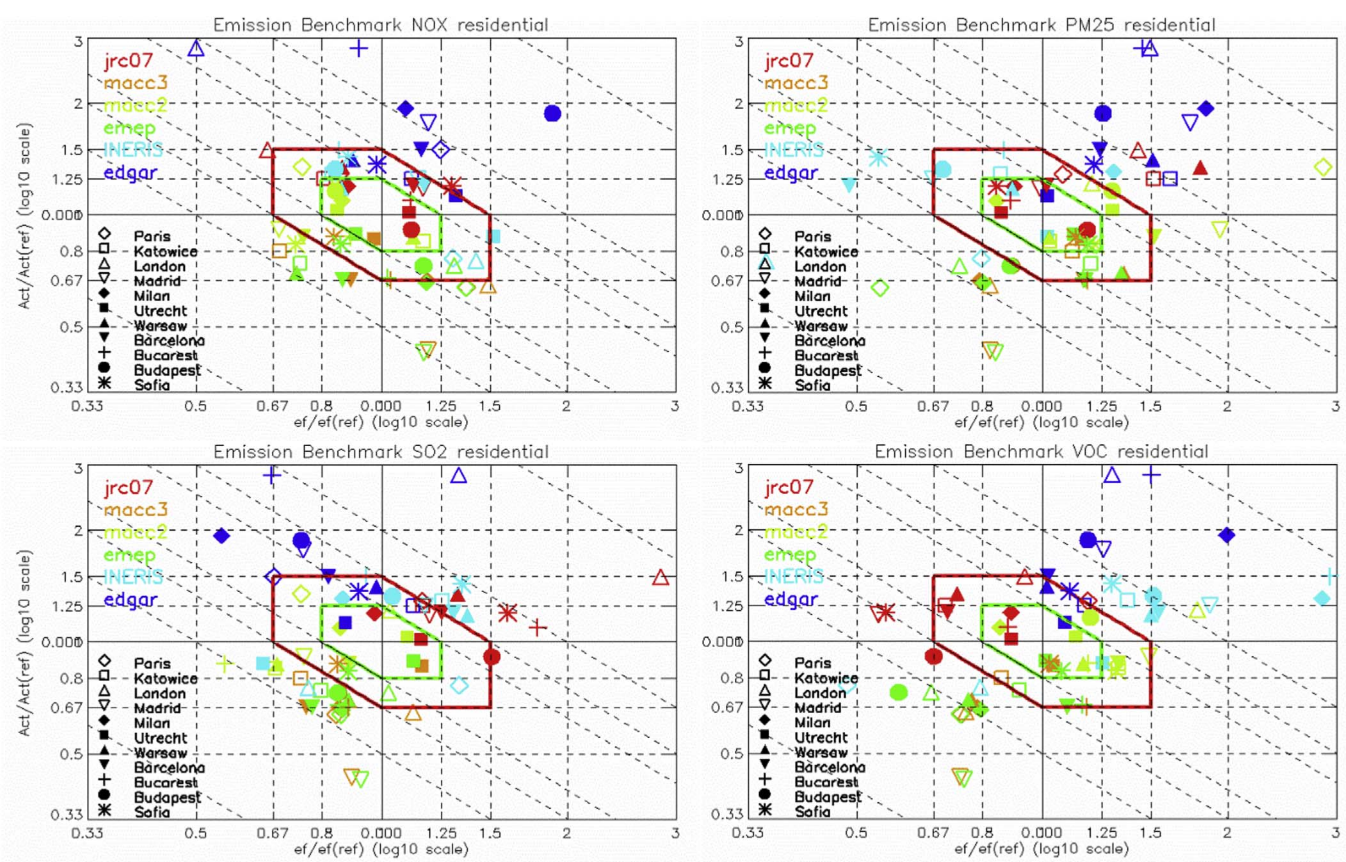

Fig. 7. Comparison between inventories using the diamond approach for the Residential Combustion sector.

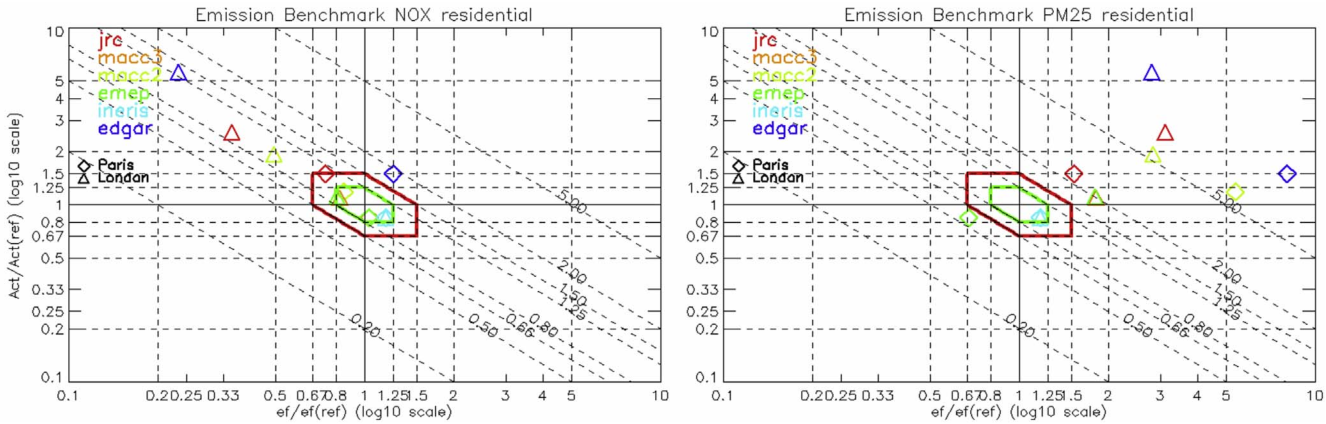

Fig. 8. Comparison between NO_x and $PPM_{2.5}$ inventories using the diamond approach for the residential sector with INERIS as a reference.

considered as a reference inventory for France and UK since the national emission inventories are directly introduced as spatial proxies (1 km^2 resolution) in the European emission dataset. In this section, assuming that the national bottom-up inventories are supposed to be the most accurate, the terms "over"- or "under-estimation" compared to this new reference can then be used for London and Paris. Fig. 8 provides a rather different picture of the spread of emissions from the residential sectors for both NO_x and $PPM_{2.5}$ compared to Fig. 7. For NO_x, the differences in terms of activities and share for JRC07 and EDGAR in London are emphasised and become much bigger, while the MACCIII ones are now more similar to the reference inventory. For $PPM_{2.5}$, in Paris the patterns are similar to those shown before, while the results are more diverse for London. Here, MACCII and JRC show larger differences from the reference inventory than before with problems of both activity levels and activity shares, while EDGAR, MACCIII and EMEP are closer to the activity levels of the reference inventory.

Industrial Sector – This sector is the one that needs the biggest efforts and improvements. In particular, EDGAR consistently has higher emissions from this sector, while INERIS and EMEP assign lower values (Fig. 9). There are in general differences between all the inventories and for all the pollutants that appear to be due to discrepancies both in terms of activity levels and shares, as indicated by the wide horizontal and vertical spreads of the points in Fig. 9.

Being largely based on Large Point Source (LPS) information, the differences seen in this industrial sector probably result mainly from differences in the choice of the relevant databases, reporting location and 'weight' of the facilities: as reported by Wang et al. (2012), the spatial accuracy of the LPS information can significantly affect the accuracy of the associated chemical transport models. The LPS databases differ in terms of spatial accuracy and thematic details (capacity or size of the single emitting facility) which strongly affect the resulting spatial variability, often combined with other inconsistencies with larger consequences (opening and closure of facilities and regular updates of the underlying databases) (Janssens-Maenhout et al., 2015; Ferreira et al., 2013). All the considered inventories rely on different versions of the E-PRTR database. The industrial emissions that cannot be linked to a specific LPS facility (i.e. diffuse fraction) since they are often below the threshold of individual facility reporting (e.g. to E-PRTR, http://prtr.ec.europa.eu) are included in this sector. Especially for small countries, the existence of threshold makes the PRTR dataset less valuable and it requires additional data for point sources falling below the threshold. Hence, the diffuse fraction has to be spatially allocated according to different proxies that may greatly contribute to the inconsistencies among inventories (Table 1). As already pointed out in section 3.2.1, in TNO-MACCII for example, emissions from SNAP34 are distributed based on the E-PRTR database, their TNO internal LPS database and on population distribution in the case of the diffuse fraction. The TNO-MACCIII introduces an improvement in the distribution of this part of the industrial sector emissions that cannot be represented by point sources (LPS). This improvement can be observed in Fig. 9 when comparing MACCII and MACCIII and will avoid a likely over-allocation of industrial emissions in urban areas. An alternative choice is the one of the EDGAR inventory which doesn't define any share of diffuse industrial emission but the whole national total is assigned to point sources.

To summarise, of the three considered sectors, road transport is the most robust, with inconsistencies mostly on activities for EDGAR and, to a minor extent, INERIS. This sector has also been reported by López-Aparicio et al. (2017) as the most consistent although, when comparing it with bottom-up approaches, all considered inventories showed underestimation of NO_x and PPM_{10} emissions. As stated in this same paper, non-exhaust emissions due to resuspension are the main reason of discrepancies for PPM10, whereas the disaggregation of traffic emissions in urban areas based on population may entail lower activity and the subsequent underestimation of NO_x emissions.

The other two sectors, in particular the industrial sector, highlight problems with both activity levels and activity shares. It is also interesting to note that in general the problems are similar for all cities in each inventory. This might mean that specific parameters of each urban area, such as land use, population density and degree of urbanization, play an important role in emission distribution.

3.3.3. Analysis in terms of pollutants

If we sum-up the emissions from the three sectors and use the diamond approach, we observe greater consistency between the inventories for all pollutants than for single macro sectors (Fig. 10). This consistency results from the compensation effects of higher and lower estimations in the individual macro-sectors. This is particularly notable for the EDGAR inventory, where the estimates of traffic emissions, which are lower when compared to the other datasets, are compensated by higher ones from the industrial and residential sectors.

The largest consistencies are mostly observed for NO_x and VOC and the lowest for $PPM_{2.5}$ and SO_2. For SO_2, the discrepancy mostly lies in the sectorial share as indicated by the large horizontal spread. It is interesting to note the differences for SO_2 between MACCII and MACCIII which are important in cities like Budapest, but small in others like Paris. These differences can be attributed to changes in the proxies used to distribute industrial emissions resulting in differences in terms of share. The proxies for industrial activities were in fact a specific target of the upgrade to MACCIII: as previously indicated, diffusive industrial emissions are allocated based on population in MACCII whereas industrial land use is used in MACCIII.

3.3.4. Uncertainties of emissions at urban scale

In this section we quantitatively summarise the results described previously. For this purpose, an estimate of the standard uncertainty is

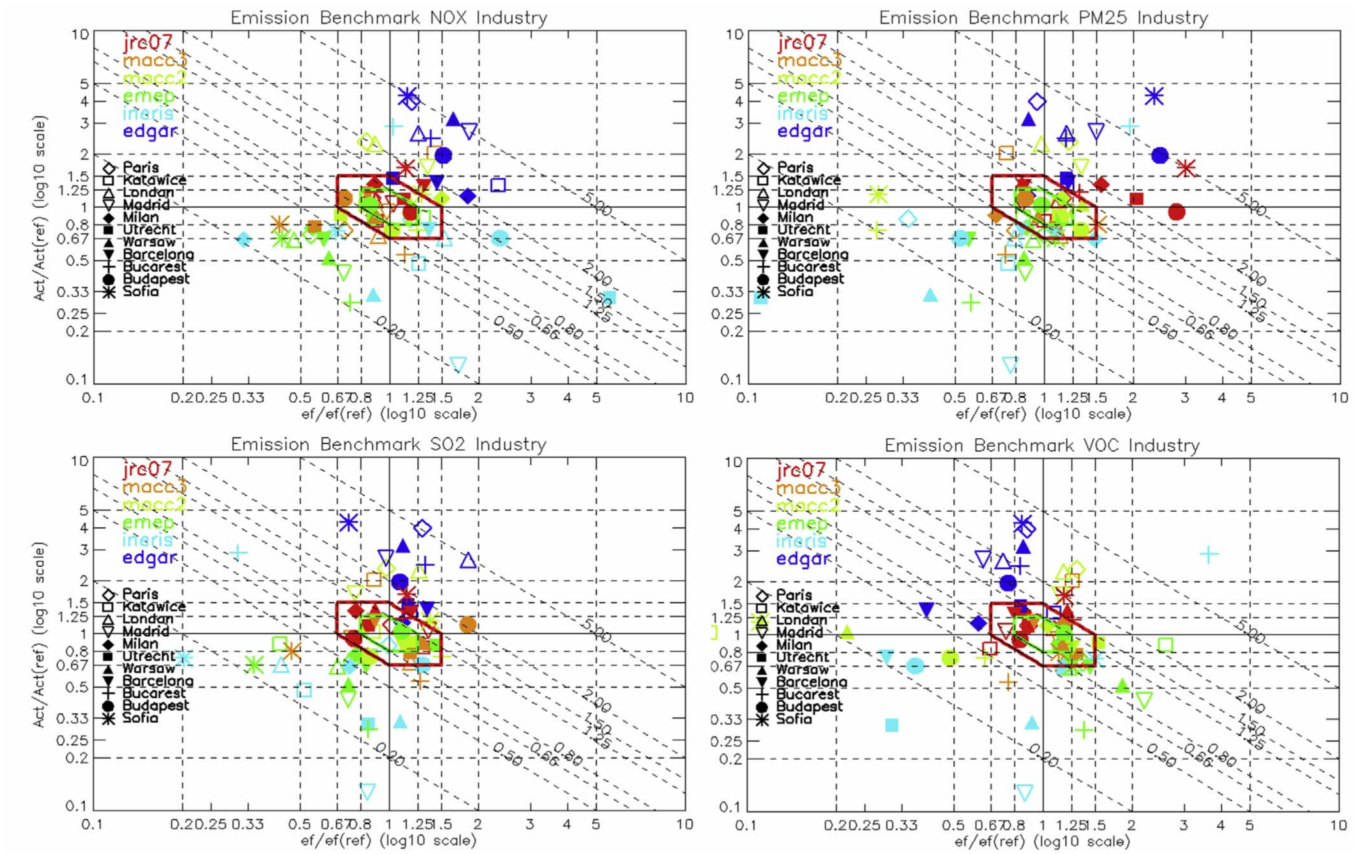

Fig. 9. Comparison between inventories using the diamond approach for the Industrial sector.

calculated for each pollutant and each sector for the 6 emission inventories. The relative standard uncertainties (u) for each city are calculated for each pollutant "p" and sector "s" according to the following formula:

$$u(p,\, s) = \frac{t^{(n-1)} * \sigma}{E_{p,s}^{mean} \sqrt{n}}$$

where $E_{p,s}^{mean}$ is the mean of the 6 emission values for a given sector and pollutant, "$t^{(n-1)}$" represents the Student's t-test probability value corresponding to a 95% confidence level and n is the number of available inventories (n = 6). The uncertainties apply to the emissions at urban scale since the starting point of all emission inventories is the national emissions total, which is identical for all of them. The uncertainties therefore reflect the expected variations resulting from the application of different spatial proxies to allocate the emissions in the urban environment.

Fig. 11 shows uncertainties up to 100% (and over this threshold for VOC) for the residential and industrial sectors whereas uncertainties of ~25% are found in the transport sector. These remarkably small uncertainties for the road transport are due to the consistency among the inventories observed in the previous paragraphs and explained by the usage of very similar spatial proxies.

It has also to be noticed that the uncertainty is very low and similar for all pollutants, including $PPM_{2.5}$ and VOC which, differently from NOx and SOx, have a significant contribution from non-exhaust emissions. This entails that even if non-exhaust emissions from resuspension is a major source of uncertainty in national inventories, they are spatially distributed in the same way by the different dataset we analysed.

Furthermore, it is also interesting to note the overall good agreement for Utrecht and Barcelona. When looking at the combination of all the considered sectors, uncertainties are generally reduced for all cities and pollutants, due to a compensation effect, although they are still very high for SO_2 and some cities such as Paris, Bucharest, Budapest and Sofia.

4. Conclusions

With the analysis presented in this paper we introduce an innovative approach for the spatial analysis of proxy-based emission inventories gridded at the European scale (i.e. ~7–10 km resolution).

Several emission inventories are available for Europe, differing substantially in terms of total emissions, sectorial emission shares and spatial distribution. It should be noted that total emissions per major sector and country can be different in the existing inventories even when based on the country reported data to CLRTAP. Since reporting takes place on an annual basis, emissions are reported annually for every historical year back to 1990. When methodological changes are made in the countries' inventory, these changes are implemented for all historical years and this may lead to significant changes in historical emission estimates. A key example is residential combustion (SNAP02) where country reported $PPM_{2.5}$ emissions for the EU28 have increased by more than 20% between 2013 and 2016 reporting. The changes are due to the differences in measurement techniques to quantify PPM emissions from small combustion installations and the lack of a clear definition for the basis on which PPM emissions should be reported.

Hence, while for the most important cities, bottom-up inventories often do exist providing more accurate information at a higher spatial resolution, for extensive air quality modelling it is still of utmost importance to be able to rely on consistent and harmonised European-wide inventories. In order to assess the potential impact of the choice of a specific inventory for air quality modelling, we analysed their spatial patterns of behaviour looking at representative urban areas over Europe.

A distinctive outcome of the work presented in this paper is the

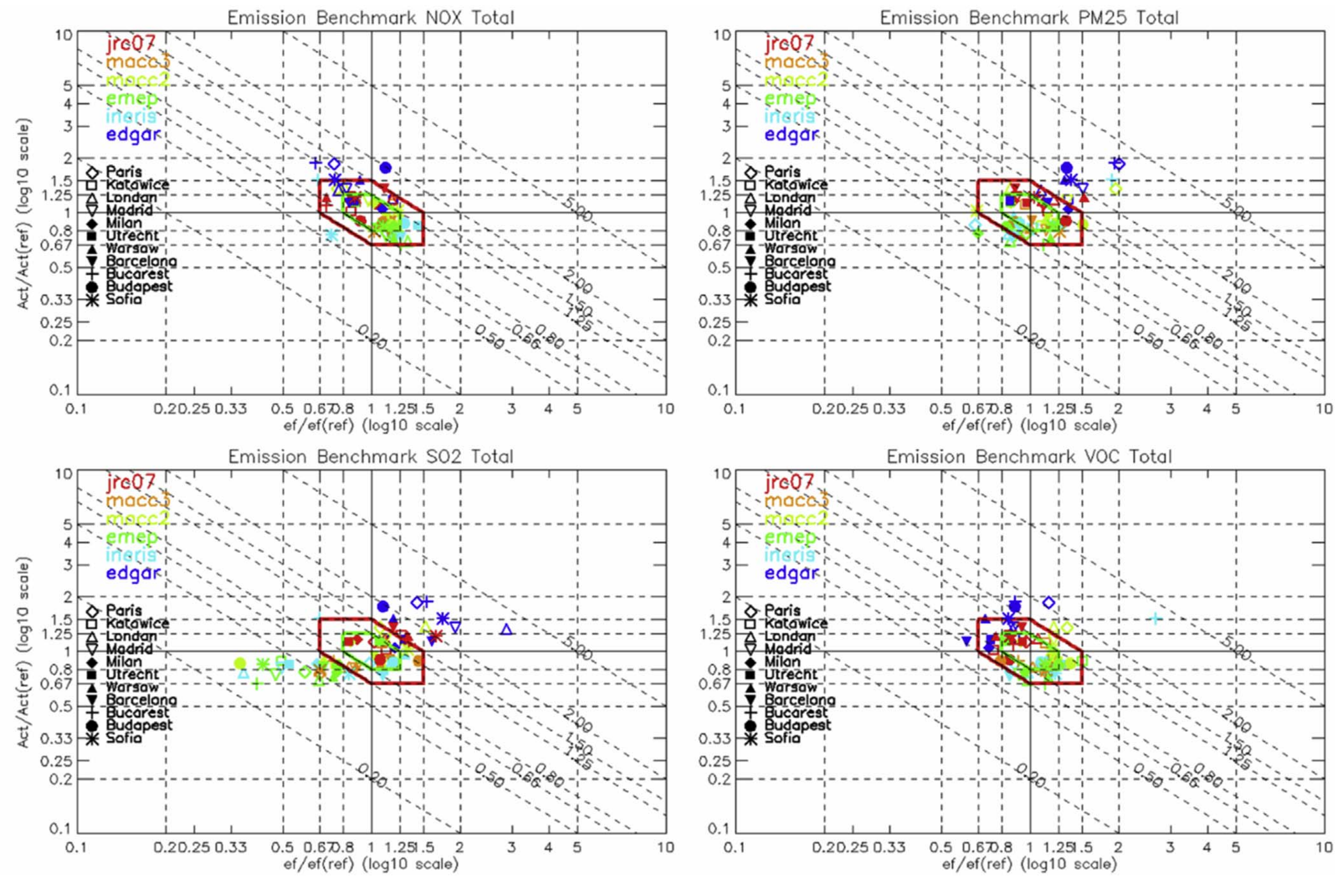

Fig. 10. Comparison between inventories using the diamond approach for the total emissions from all the considered sectors.

significant difference between regional emission inventories due to the choices made in terms of disaggregation approach and selection of spatial proxies. Moreover, our study underlines those sectors where additional efforts are needed in the framework of regional air quality assessments.

For all inventories, it appears necessary to review, compare and develop new methodologies and proxies for the spatial disaggregation of emissions from the industrial sector. The large inconsistencies observed may be in part due to the different methodologies and assumptions used to allocate the diffuse industrial emissions. Emissions from Medium Combustion Plants (MCP, > 1 MWth and < 50 MWth) are just now starting to be regulated by the EC (Directive, 2015/2 193) and consequently no information on these facilities (e.g. geographic location, emissions) is available. Considering that the number of MCP in the EU is estimated to be around 143,000 (European Union, 2016b), having detailed information of these facilities would improve the allocation of industrial emissions and reduce the observed discrepancies.

As it was previously highlighted (e.g. Guevara et al., 2014; López-Aparicio et al., 2017), the use of the population density as proxy to allocate the diffuse fraction of industrial emissions results in an over-allocation of emissions in urban areas (e.g. TNO-MACCII). Even though the distribution of diffuse emission based on land use cover data is an improvement (e.g. TNO-MACCIII), this approach still needs further development. The main reason is that the land use classification includes in the 'industry' class areas that are commercial rather than industrial. It is also important that inventories base the distribution of emissions from Large Point Sources emissions on the latest available LPS dataset and that, for the sources below the emission thresholds which can be very important especially in small countries, appropriate complementary dataset are adopted.

Particular attention should also be given to the residential sector, if possible comparing bottom-up estimates to better calibrate the spatial patterns of emissions from wood and coal burning, in order to reflect the significant variations between countries. Furthermore, city-specific features such as district heating should be taken into account; in these cases, a much lower share of residential emissions would be expected over the city compared to individual heating sites. At the same time, the traditional proxies used for gridding residential emissions (e.g. population density) would not be any more relevant.

Based on the differences highlighted in this analysis, we list the main aspects for each inventory that could be important to review. It has to be noted that these issues have been identified by a comparison between gridded Top-down inventories and, since there is no way to directly verify the results of the disaggregation, they have to be considered as hints for a critical revision of the chosen downscaling methodologies. Considering that none of the analysed inventories can be considered as a true reference, it is also important to emphasise that the consensus found for certain sectors/pollutants (e.g. NO_x traffic emissions) does not necessarily indicate that the uncertainty in the emission inventories is low. A high level of consensus may be due to similar assumptions used in all the inventories or similar sources of uncertainties (e.g. laboratory versus on-road traffic emission factors, Degraeuwe and Weiss, 2017):

EDGAR: The importance of residential and road traffic emissions appears to be systematically estimated as lower (SNAP02) and higher (SNAP07) over urban areas (the split of national totals in terms of macro-sectors seem to be in line with the other inventories).

This emission inventory is generally the one that presents the largest inconsistencies when compared to the other analysed emission inventories. At the same time, it is the only one that offers a global spatial coverage, hence dealing with a wider range of data sources which need to ensure consistency and representativeness for the different parts of the globe. This fact indicates that when working at different scales, the availability and detail of spatial proxies may change.

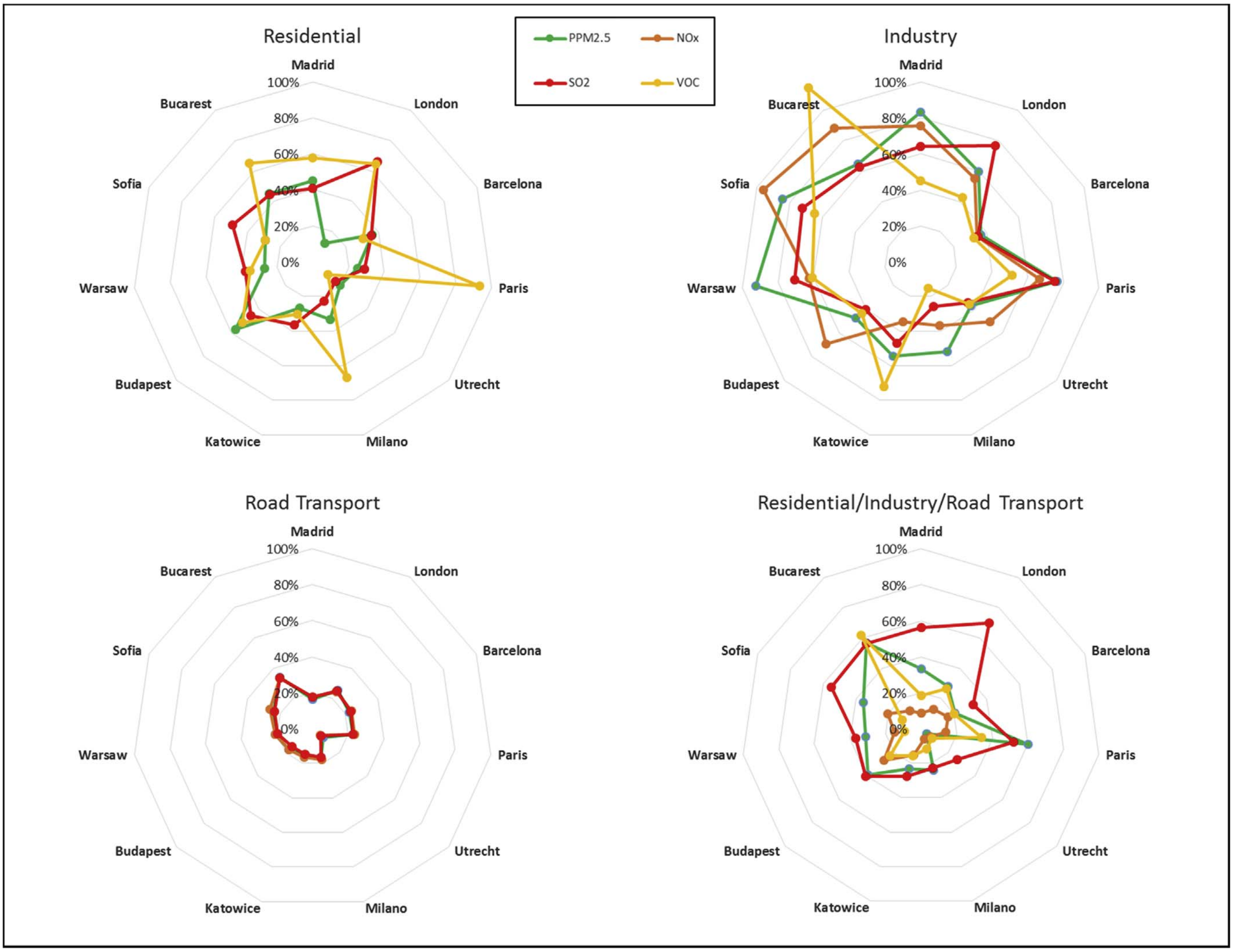

Fig. 11. Percentage standard uncertainties for the transport, residential and industry sectors for the 11 selected cities. VOC for SNAP07 and NO_x for SNAP02 are not shown here, since, for these pollutants, the contribution from these sectors to the total emission is less than 10% (average value of all considered inventories).

INERIS: The spatial disaggregation of emissions from on-road traffic should be checked for some eastern cities (Bucharest, Sofia) for which much lower values are reported.

MACCII-MACCIII: As expected, a general improvement from MACCII to MACCIII is observed with very large changes for some of the cities. In general, in MACCIII, industrial and residential emissions are now distributed more outside of the city domains and less within the urban areas (Kuenen et al., 2014). For the industrial sector, the area sources which were distributed using population density in MACCII are gridded over the industrial land use area in MACCIII.

For the residential sector, MACCIII assigns lower amounts of wood or coal burning to the city centers. The estimates for wood combustion and the spatial distribution have been revised; for Eastern European countries, the emissions from this source have been significantly increased.

EMEP Industrial emissions proxies and methodologies should be checked since for all pollutants much lower values than the other inventories are reported.

*JRC*07 Particular attention should be given to residential emissions in Eastern Europe (in particular Poland); the country inter-variability of urban residential emissions (Wood and Coal burning) has not been properly addressed.

These first results provide an insight for the identification of the main issues and differences among the emission inventories commonly used at the European scale for air quality modelling and some recommendations are provided with the aim of working towards the harmonisation of spatial downscaling and proxy calibration, in particular for policy purposes.

Further work will be needed in order to provide a deeper insight into emission spatial patterns through a comparison at a finer scale with local bottom-up inventories, which rely on massive and detailed spatial information such as point sources, detailed censuses and traffic statistics or, as alternative, with the national grids at 0.1*0.1° resolution recently reported to EMEP by many European countries. Such a comparison would help calibrate proxies at a regional/local scale rather than using common ones for such diverse and extended areas.

Finally, and considering that one of the main aims of the analysed inventories is to provide emission inputs for air quality modelling, future work should also consider the influence of uncertainties in proxy-based emission inventories when they are used in atmospheric chemistry models.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2017.10.032.

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