

# On the validity of the incremental approach to estimate the impact of cities on air quality

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## ABSTRACT

The question of how much cities are the sources of their own air pollution is not only theoretical as it is critical to the design of effective strategies for urban air quality planning. In this work, we assess the validity of the commonly used incremental approach to estimate the likely impact of cities on their air pollution. With the incremental approach, the city impact (i.e. the concentration change generated by the city emissions) is estimated as the concentration difference between a rural background and an urban background location, also known as the urban increment. We show that the city impact is in reality made up of the urban increment and two additional components and consequently two assumptions need to be fulfilled for the urban increment to be representative of the urban impact. The first assumption is that the rural background location is not influenced by emissions from within the city whereas the second requires that background concentration levels, obtained with zero city emissions, are equal at both locations. Because the urban impact is not measurable, the SHERPA modelling approach, based on a full air quality modelling system, is used in this work to assess the validity of these assumptions for some European cities. Results indicate that for PM<sub>2.5</sub>, these two assumptions are far from being fulfilled for many large or medium city sizes. For this type of cities, urban increments are largely underestimating city impacts. Although results are in better agreement for NO<sub>2</sub>, similar issues are met. In many situations the incremental approach is therefore not an adequate estimate of the urban impact on air pollution. This poses issues in terms of interpretation when these increments are used to define strategic options in terms of air quality planning. We finally illustrate the interest of comparing modelled and measured increments to improve our confidence in the model results.

## 1. Introduction

Most European cities suffer from poor air quality and regularly exceed both the thresholds prescribed by the Air Quality Directive (EEA, 2016) and guidelines recommended by the World Health Organization (WHO). This is particularly the case for fine particulate matter (PM<sub>10</sub>) for which both the daily (50 µg/m<sup>3</sup> not to be exceeded more than 35 days a year) and yearly average limit values (40 µg/m<sup>3</sup>) are regularly exceeded in many cities and several regions in Europe. Similar conclusions hold for PM<sub>2.5</sub> where few cities manage to keep concentrations below the levels recommended by the WHO (10 µg/m<sup>3</sup> on an annual basis). Adverse health effects and premature deaths are two of the major impacts resulting from poor air quality and an average life loss of about 8–10 months is the current estimate for the most polluted European regions (Southern Poland, Po Valley, Benelux ...) and cities, consequently air pollution remains the single largest environmental health risk in Europe according to WHO (WHO Regional Office for Europe and OECD, 2015).

Actions have been proposed and taken at the international (e.g. Amann et al., 2011), national (e.g. D'Elia et al., 2009) and/or local scale (e.g. Giannouli et al., 2011) to reduce air pollution. Although those

actions have resulted in an overall improvement of the air quality over the years (Maas and Grennfelt, 2016), problems remain, although they tend to be localised in specific regions and many cities. A key issue is thus to determine at which of the above scales to implement air quality policy actions in order to abate air pollution most effectively. Central to this for cities, is a quantitative assessment of the different contributions to air pollution in the city (city, regional, national and international). In this work, we review two main approaches that exist to quantify the impact of a city on its air pollution levels:

The first approach is based on spatial concentration increments. Two background locations (city and rural) are compared: the city location is assumed to be representative of the urban background levels while the rural location is assumed to be representative of the levels far away from the city. The difference between the concentrations at these two locations gives the impact of a city on its own air pollution. Because this approach was proposed by Lenschow et al. (2001) to quantify the impact of the city of Berlin on its air pollution, we will henceforth refer to it as the "Lenschow" approach. It may be applied with either measurements or modelling results (e.g. Squizzato and Masiol, 2015; Timmermans et al., 2013; Petetin et al., 2014; Pey et al., 2010; Ortiz and Friedrich, 2013; Keukens et al., 2013). Recently, Kiesewetter and

Amann (2014) combined a measured-based incremental approach with modelling results over the entire European domain in the frame of the Clean Air for Europe Programme. In this mixed approach, urban impacts are based on observed pairs (spatial increments) but are corrected with modelling results to ensure that (1) all secondary particulate matter is given a regional origin and (2) the resolution-increment (i.e. the difference between concentrations modelled at 7 and 28 km spatial resolutions) is given a city origin.

The second approach is based on Chemistry Transport Models (CTM) that can be used to quantify the impact of cities on their pollution by performing coupled base case simulations and sensitivity (or brute force) simulations where city emissions are set to zero (Pecorari et al., 2012; Chen et al., 2017; Guo et al., 2016; Sciare et al., 2010). Here the city impact is also obtained by a simple difference between the two simulation results, within the city. Because air quality models typically require intensive CPU resources, this approach maybe too complex for a generalization to many cities. Unlike the Lenschow incremental approach, this methodology is limited to models and is henceforth referred to the “CTM urban impact”.

In this work, we first examine the theory behind the calculation of the urban impact and review the assumptions made when using the two approaches mentioned above, the Lenschow urban increment and the CTM urban impact. Model results obtained for a few European cities are then used to illustrate the differences between the approaches and examine their limitations. The focus is on PM<sub>2.5</sub> as it is responsible for most of the health related impacts, but results for NO<sub>2</sub> are also discussed.

## 2. Urban impact and urban increment

The urban impact ( $I$ ) is a theoretical construct that is defined as the change of concentration in a city due to the emissions coming from the city itself. The easiest way to express this is by imagining that all the emissions from within a city, or zone within a city are set to zero. Thus at a city location (city superscript)  $I$  is defined as:

$$I_{cf}^{city} = C^{city} - B_{cf}^{city} \quad (1)$$

where  $C^{city}$  is the concentration level reached when both in-city and extra-city emissions are active and  $B_{cf}^{city}$  is the background concentration level reached when emissions are set to zero for a fraction of the city ( $cf$ ). The city fraction is used to distinguish different zones that can be attributed to the same city (e.g. Greater or inner London). The urban impact ( $I_{cf}^{city}$ ) and the background ( $B_{cf}^{city}$ ) explicitly depend on the size of the city fraction, over which emissions are switched off. All terms in (1) and the following relations use superscripts to indicate the location where the concentration is analyzed (receptor) and subscripts to indicate the area over which emissions are switched off (source). Relation (1) can similarly be applied to any rural location (“rur” superscript) at a given distance (d) from the city centre to give the impact of emissions from the city on the rural background location:

$$I_{cf}^{rur}(d) = C^{rur}(d) - B_{cf}^{rur}(d) \quad (2)$$

The concentration at any location ( $C^{rur}(d)$ ,  $C^{city}$ ) is then the sum of two components: an urban impact ( $I_{cf}^{rur}(d)$ ,  $I_{cf}^{city}$ ) that represents the concentration due to the in-city emissions and a background level ( $B_{cf}^{rur}(d)$ ,  $B_{cf}^{city}$ ) that represents the remaining concentration when in-city emissions are set to zero (Fig. 1).

The Lenschow urban increment and the urban impact can be connected by differencing (1) and (2):

$$I_{cf}^{city} = \underbrace{(C^{city} - C^{rur}(d))}_{\text{Lenschow increment}} + \underbrace{I_{cf}^{rur}(d)}_{\text{City spread}} + \underbrace{(B_{cf}^{rur}(d) - B_{cf}^{city})}_{\text{Background deviation}} \quad (3)$$

According to relation (3), the urban impact at the city centre is the sum of three components:

- The Lenschow urban increment that corresponds to the concentration difference between the city centre and the rural background locations. The importance of this component depends on the distance (d) at which the rural background location is selected.
- The “city spread” that quantifies the impact of the city at the rural background location. It is equal to the urban impact at the rural background location. This component depends on distance (d) and on the size of the city fraction considered.
- The “background deviation” that quantifies any concentration difference between the city and rural background locations when the in-city emissions are set to zero. This component also depends on distance (d) and on the size of the city fraction considered.

In equation (1),  $B_{cf}^{city}$  cannot be measured because this would require measurements to be taken with in-city emissions set to zero. Two main options exist to approximate its value. With the “Lenschow increment” approach  $B_{cf}^{city}$  is assumed to be equal to the background rural concentration measured away (at distance “d”) from the city, i.e.:

$$B_{cf}^{city} = C^{rur}(d) \quad (4)$$

whereas the “CTM scenario” directly quantifies  $B_{cf}^{city}$  by performing a dedicated model simulation in which city emissions are switched off. Equation (3) shows that the Lenschow urban increment approximates to the urban impact when the sum of the city spread and the background deviation is negligible. These two terms can therefore either be negligible each or compensate each other. As shown afterwards, compensation between these two terms may coincidentally occur for PM<sub>2.5</sub> or NO<sub>2</sub>, but this would happen when a limited fraction of the city is considered. Because the objective is to assess the full impact of given city, each of the two following assumptions need to be fulfilled individually.

**Assumption I.** The urban impact at the rural site is negligible.

$$I_{cf}^{rur}(d) \cong 0 \quad (5)$$

This assumption implies that the rural location is located far enough from the city not to be subject to its’ influence.

**Assumption II.** The background levels are spatially homogeneous.

$$B_{cf}^{rur}(d) \cong B_{cf}^{city} \quad (6)$$

This assumption implies that both locations should not be too far away from each other, which is counter to **Assumption I**.

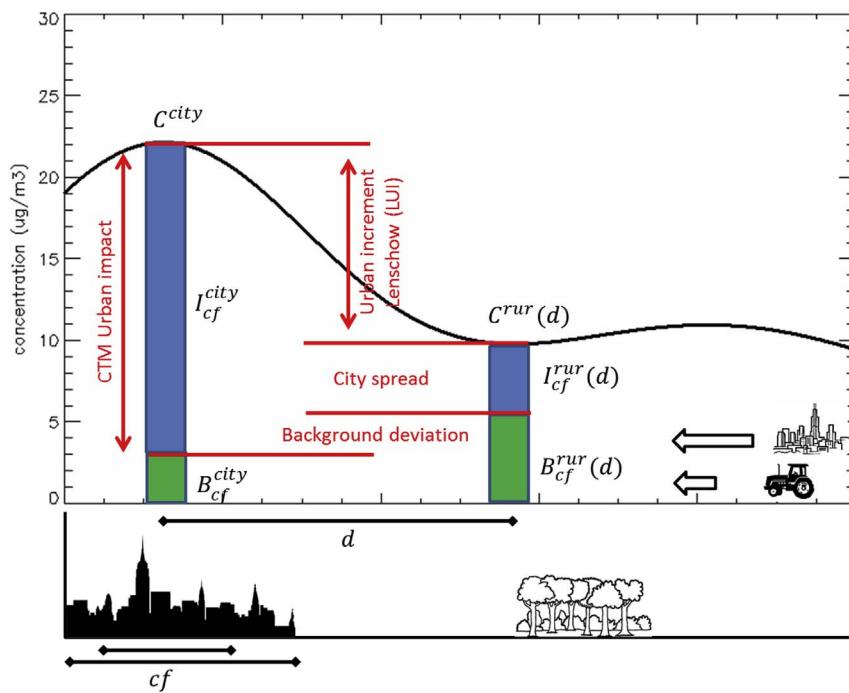
While Lenschow et al. (2001) summarize (5) and (6) by assuming that the differences between the urban background and the rural background locations can be attributed to the emission sources within the city area, Pey et al. (2010) note that the rural background location may be affected by urban pollution, and consequently the net urban impacts presented in their work should be considered as minimum. Although the above-cited works mention underlying assumptions, no quantification is proposed.

Given that, it is not possible to measure directly the urban impact, we use in this work a modelling approach to quantify all terms in equation (3) for some cities in Europe. We will consider the CTM urban impact as the reference and assess how well the Lenschow urban increment approximates it for those cities, for different pollutants. We will also examine the behaviour of the Lenschow increment, the city spread and the background deviation for different choices of the city fraction for the selected cities.

## 3. Comparison for selected European cities

### 3.1. Methodology

To cope with one of the main limitations of the CTM-scenario approach, namely the computer time required to perform scenario



**Fig. 1.** Schematic visualisation of the concentration split into an urban impact and a background level at an urban (city centre, denoted with the superscript “city”) and rural site (denoted with the superscript “rur”). The CTM urban impact, the Lenschow urban increment, the city spread and the background deviation are indicated in red (see text for more details). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

simulations, the SHERPA approach (Thunis et al., 2016) is used. SHERPA is based on a set of simplified source-receptor relationships (SRR) that link emission changes to concentration changes. These SRR are based on a set of full CTM simulations (Clappier et al., 2015; Pisoni et al., 2017). SHERPA mimics the CTM responses to emission changes, but with a much shorter computing time. In the current configuration, SHERPA operates with the CHIMERE (Menut et al., 2014) model covering the whole of Europe at 7 km spatial resolution. SHERPA can thus calculate the impact of emission reductions applied over any urban, regional or national area. Schaap et al. (2015) showed that a  $7 \times 7$  km resolution was an accurate resolution to capture urban background concentrations, focus of this work. Because of its’ simplifying assumptions, SHERPA only calculates yearly average concentration levels of NO<sub>2</sub> and PM<sub>2.5</sub>. It is important to note that no measurement is available to validate CTMs for model-responses to emission changes. This is the reason why CTMs are tested regularly in the frame of inter-comparison exercises. With the SHERPA approach, the validation tests aimed at ensuring that the simplified SRR could mimic well the CTM’s behaviour. A series of validation tests in which emissions were reduced in different city and regional areas showed the ability of SHERPA to reproduce the responses of the full CHIMERE CTM model with an overall accuracy of 10%, reaching 5% at urban locations, which are the focus of this work (Thunis et al., 2016; Pisoni et al., 2017). We assume in the following that SHERPA accurately reproduces the results of the CHIMERE CTM for the scenarios used.

Three different urban fraction sizes are defined to perform our analysis. First, cities are defined according to EU-OECD (OECD, 2012) as local administrative units where the majority of the population lives in an urban centre. A larger “city” area is defined as the functional urban area (FUA) consisting of the city and its’ commuting zone, defined as the surrounding travel-to-work areas where at least 15% of the employed residents work in the city. For London and Paris a smaller urban area, referred to as the inner city is also considered (Fig. 2). Table 1 lists the population and the area associated with each of these city areas.

In the next section, we use the SHERPA approach together with these city fraction definitions to compare the Lenschow urban increment with the CTM urban impact. We first focus on PM<sub>2.5</sub> before extending the analysis to NO<sub>2</sub>. We finally discuss possible comparisons

with measurements.

### 3.2. Urban impacts for PM<sub>2.5</sub>

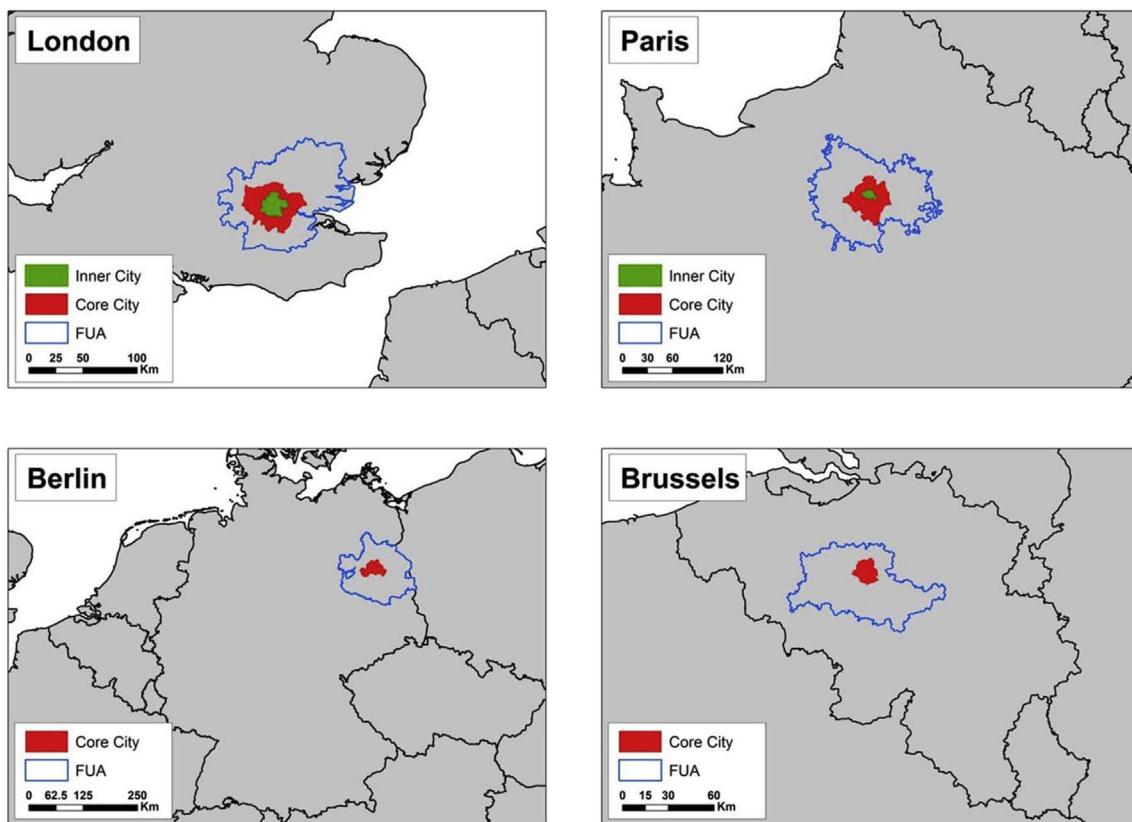
Particulate matter (PM<sub>2.5</sub>) is a pollutant that can be emitted directly (primary particulates) or be formed after a series of complex chemical formation processes (secondary particulates) (see the review by Fuzzi et al., 2015). Depending on meteorological conditions, their residence time in the atmosphere is estimated to range from several days up to one week in the lower troposphere (Jaenicke, 1980). It is worthwhile to note that even with relatively light wind speeds, these residence times imply travel distances in the lower troposphere of the order of a thousand kilometres. In addition, air masses do not always follow straight trajectories, consequently under certain circumstances primary emissions from a city may react with precursors emitted elsewhere to form secondary particles that then return to the city where they were emitted. Given this phenomenology, the two assumptions (5) and (6) imply that:

- Primary emissions (PPM) from the city do not reach the rural location and gas-phase emission precursors (e.g. NO<sub>x</sub>, SO<sub>2</sub> ...) do not form secondary aerosols that impact the rural location [[Assumption I](#)].
- PPM or gas-phase precursors emitted by other cities or by activity sectors outside the city area (e.g. agriculture) influence the rural and city centre locations in the exact same way [[Assumption II](#)].

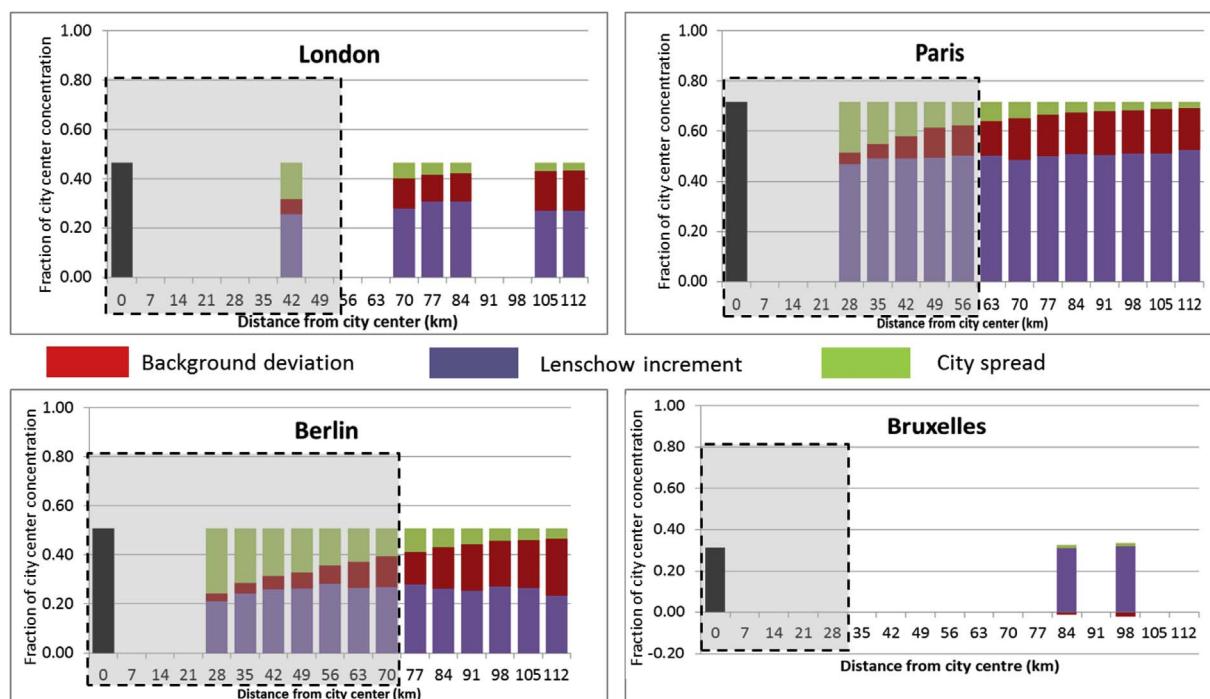
In the following section, we assess how well these two assumptions are fulfilled as a function of the city, the distance from the city centre and the definition of the city fraction.

#### 3.2.1. Functional urban areas (FUA)

In these experiments, in-city emissions are set to zero over an area corresponding to the FUA ( $cf = FUA$ ) for each of the four cities. Emissions over grid cells that are divided by the FUA boundary, are allocated proportionally to the FUA area within that cell. SHERPA is then used to quantify all terms in equation (3) with respect to distance ( $d$ ) from the city centre. Results are expressed relative to the urban background PM<sub>2.5</sub> concentration modelled at the city centre (indicated



**Fig. 2.** Overview of the urban cores (in red), functional urban areas – FUA (in blue) and inner city (in green) areas for Berlin, Brussels, London and Paris, analyzed in this work. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 3.** Quantification of the modelled Lenschow increment (purple), modelled city spread (green) and modelled background deviation (red) for all four cities, as a function of distance ( $d$ ) from the city centre. Urban emissions are reduced over each FUA of which the average radius is indicated by the shaded rectangle. The black column represents the modelled urban impact. All results are expressed in percentage of the city centre modelled PM<sub>2.5</sub> concentrations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

**Table 1**

Characteristics (population (pop -  $10^6$  inhabitants) and area covered (area -  $10^3 \text{ km}^2$ )) for the 4 cities analyzed. The equivalent radius (Eq. Rad - km) column indicates the average radius of the city that is used in the next sections to delineate the city dimension in Fig. 3 and following.

	Extended city (FUA)			Urban Core			Inner-City		
	pop	area	Eq. Rad.	Pop	area	Eq. Rad.	pop	area	Eq. Rad.
London	11.76	8,024	51	7.83	1,576	22	3.0	318	10
Berlin	4.92	17,484	75	3.54	1,079	19			
Brussels	2.42	3,266	32	1.07	162	7			
Paris	12.17	12,098	62	9.37	1,602	23	2.2	105	6

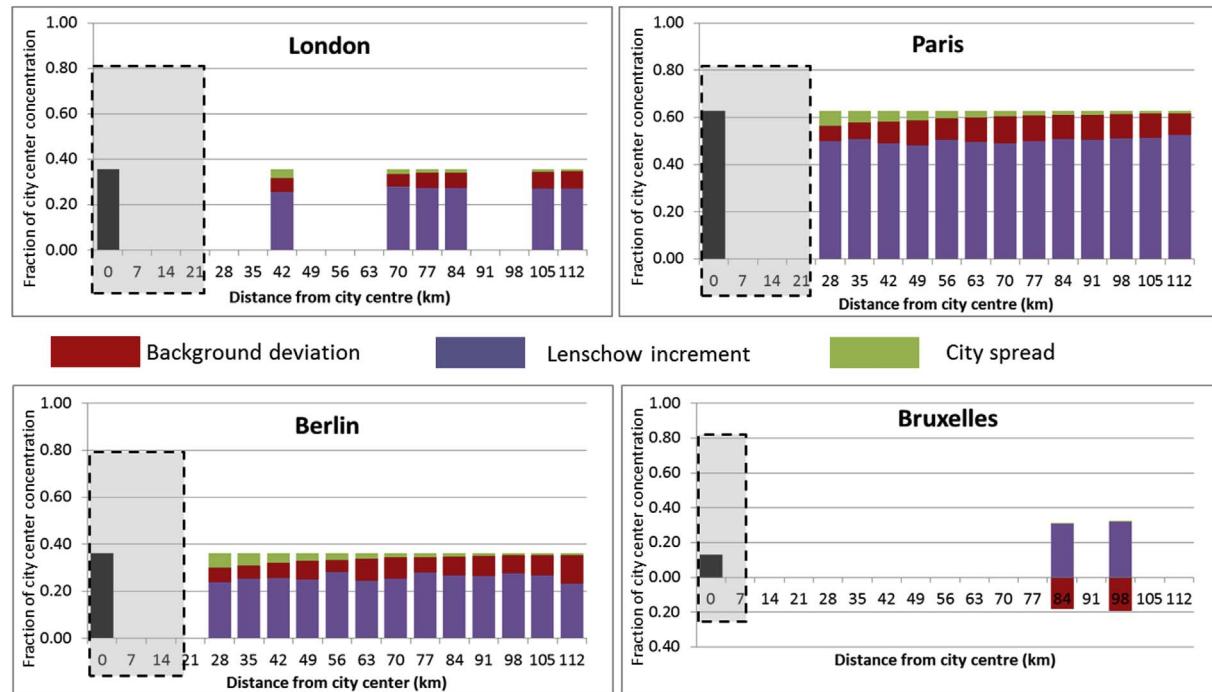


Fig. 4. Same as Fig. 3 but for cities limited to their urban core fractions (see text and Table 1 for details).

as fractions in Fig. 3 and following). The first column (black) in Fig. 3 represents the CTM urban impact ( $I_{FUA}^{\text{city}}$ ), calculated as the concentration change (at the city centre) between two simulations, one with and the other without the FUA emissions. The purple, green and red fractions represent the Lenschow urban increment ( $C^{\text{city}} - C^{\text{rur}}(d)$ ), the city spread ( $I_{FUA}^{\text{ur}}(d)$ ) and the background deviation ( $B_{FUA}^{\text{ur}}(d) - B_{FUA}^{\text{city}}$ ), respectively, as a function of the distance from the city centre ( $d$ ). The Lenschow urban increment is calculated directly from the base-case concentration fields (difference between the city and rural background locations). The background deviation is obtained similarly but from the no-city emission simulation and the city spread is obtained by difference between the base-case and the latter scenario. The three terms are calculated along the four main geographical axes (East, West, North and South) on a grid-cell basis, at 7 km spatial resolution. To ensure that only rural grid cells are considered for estimating the rural concentration, a simple constraint is imposed on grid cell emissions. Only those cells where PPM emissions are less than 5% (level chosen arbitrarily) of the emissions of the city centre grid-cell are kept for the analysis. The available grid cell results along the 4 directions are then averaged into a single value that is reported in Fig. 3. The dashed line rectangles indicate the approximate city fraction extension (obtained with the equivalent radius reported in Table 1) over which emissions are switched off. Finally note that columns inside the equivalent radius of the functional urban area mean that there are rural areas along the four geographical axes within the functional urban area. In practice, the incremental approach would only consider rural sites that are outside

the city boundaries.

As seen from Fig. 3, the city spread component decreases with distance but this decrease is slow and significant city spreads are modelled far away from the city. This is most likely a consequence of the complex PM formation processes mentioned above that result in long residence times in the atmosphere. For results that approximate to a negligible city spread and fulfil the first assumption, the rural location should be located at least 80–100 kms from the city centre. At those distances, however, the background deviation becomes important and assumption II of background homogeneity is no longer valid. This background deviation is due to the impact of other (than the city) emission sources (e.g. other cities, agriculture, industries) that start playing an important role when moving away from the city centre. The background deviation reaches around 20% of the urban impact (black column) for Paris and London and 50% for Berlin.

It is therefore impossible when considering the entire functional urban area to find rural locations that satisfy both assumptions simultaneously, i.e. a location for which the Lenschow urban increment is representative of the CTM urban impact. For Brussels, the smallest city in our examples, the Lenschow urban increment underestimates the CTM urban impact, regardless of the distance from the city centre. This underestimation is around 5% in Brussels, 25% in Paris, 40% in London and 50% in Berlin.

For a smaller city like Brussels, the spatial emission gradients between the city and its neighbouring rural locations are relatively weak and we find only a few grid cells that fulfil the “rural” emission

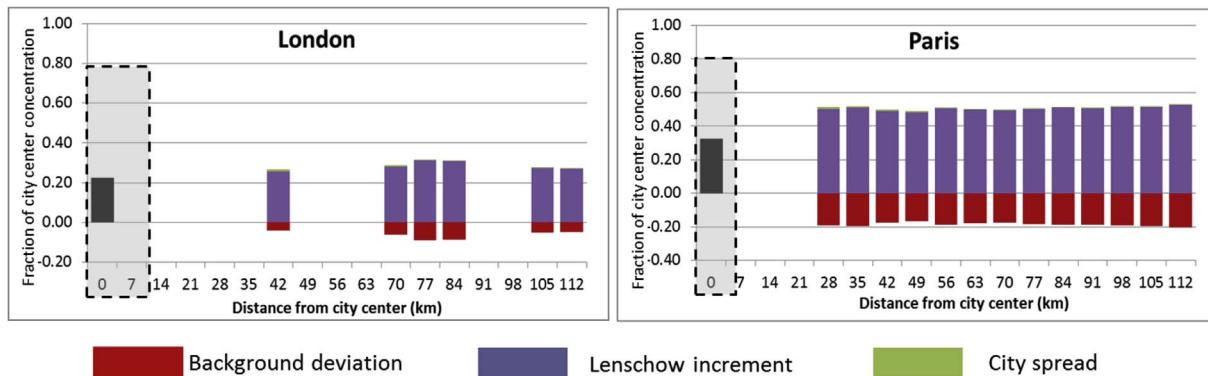


Fig. 5. Same as Fig. 3 but for city areas (area over which emissions are switched off) limited to the inner city areas for London and Paris (see text and Table 1 for details).

constraint (as seen from Fig. 3 where only few cells are represented). However, the Lenschow increment closely approximates the urban impact for those few cells located far away from the city (beyond 80 km).

In the next section, we repeat the analysis for the same cities but for using the urban core and inner city as the city definition.

### 3.2.2. Urban core and inner-cities

Fig. 4 provides similar results to Fig. 3 but with emissions set to zero over an area corresponding to the urban cores (see Table 1 for details). For London and Paris, an additional option is investigated with emissions set to zero in the inner city only (Fig. 5). Note that given the limited number of grid cells considered for this latter city size with a 7 km resolution, results are likely less robust for the inner city than for larger city sizes. The Lenschow urban increment remains unchanged regardless of urban size because it is derived from spatial concentration differences, while the city spread and background deviation both vary. For the urban core, the city spread ( $I_{Urban\ core}^{rur}(d)$ ) is as one would expect less important than for the FUA ( $I_{FUA}^{rur}(d)$ ), because the total emissions set to zero in this area is less than for the larger FUA. This component similarly decreases with distance from the city. Because of this reduced “city spread” distance, the background deviation ( $B_{Urban\ core}^{rur}(d) - B_{Urban\ core}^{city}$ ) is also less important. Consequently the Lenschow urban increment ( $C^{city} - C^{rur}(d)$ , purple fraction) gets closer to the CTM urban impact ( $I_{Urban\ core}^{city}$ , black column) than in the case of the FUA.

The Lenschow approach continues to underestimate the urban impact for the three larger cities (for similar reasons as reported for the FUA) while it significantly overestimates it in Brussels. In this latter case, this is explained by the fact that (1) the urban core is small enough to ensure that no city spread is visible and (2) the background deviation becomes negative. Because emissions are switched off only in the city core in the case described here, emissions within the commuting zone remain active. In the case of Brussels, these commuting zone emissions are large enough to influence the city centre concentration, in such a way that the resulting background concentration becomes larger than the rural background, leading to a negative background deviation.

For city sizes restricted to the urban cores, the Lenschow urban increment underestimates the urban impact by 15% in Paris, 25% in London and 35% in Berlin, whereas it overestimates it in Brussels by a factor of 2.

For London and Paris, results for an even smaller city fraction size (inner city) (see Table 1 for details) are presented in Fig. 5. As is found with the Brussels urban core above, the city spread ( $I_{Inner\ city}^{rur}(d)$ ) disappears, the background deviation ( $B_{Inner\ city}^{rur}(d) - B_{Inner\ city}^{city}$ ) is negative and the Lenschow urban increment ( $C^{city} - C^{rur}(d)$ ) overestimates the urban impact ( $I_{Inner\ city}^{city}$ ).

It is important to note that the CTM urban impact, the city spread and the background deviation depend explicitly on the definition of the

city, whereas the Lenschow urban increment does not. This is highlighted in Fig. 6 in which an overview of the urban impacts obtained for the different city definitions examined is provided. If the increase in the CTM urban increment as the city fraction increases is characteristic of cities in general, then for all cities there exists a city fraction for which the Lenschow increment equals the CTM urban impact. Unfortunately it varies from city to city: it is close to the FUA for Brussels; it lies between the urban core and the inner city for Paris and London and is smaller than the inner city for Berlin. As can be expected and is seen in Fig. 6, city fraction is a key determinant of the CTM urban impact, which varies from 30 to 70% in Paris, from 10 to 30% in Brussels, from 35 to 50% in Berlin and from 20 to 50% in London, when moving from a small to a larger city fraction. Depending on the city fraction used, cities (e.g. Paris, Brussels, London) can either be considered as net importers (city core) or net producers (FUA) of their pollution, with obvious implications in terms of policy.

This point also appears clearly from equation (3) re-written below, in which only the right-hand terms explicitly depend on the city size:

$$LUI = (C^{city} - C^{rur}(d)) = I_{cf}^{city} - I_{cf}^{rur}(d) - (B_{cf}^{rur}(d) - B_{cf}^{city}) \quad (7)$$

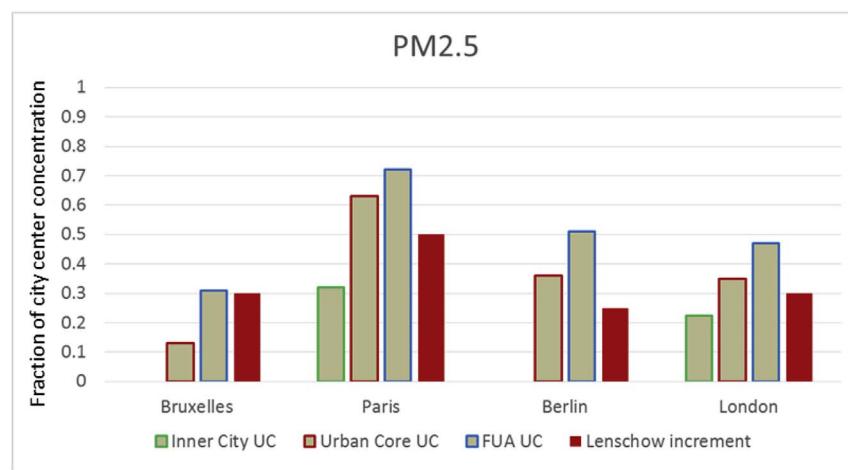
By varying the distance ( $d$ ) and/or the city fraction ( $cf$ ) in this equation, we can identify a city fraction that is implicitly associated to the Lenschow urban increment ( $LUI$ ). This implicit city fraction size is the size for which the sum of the city spread and the background deviation tends to zero:

$$I_{cf}^{rur}(d) + (B_{cf}^{rur}(d) - B_{cf}^{city}) \approx 0 \Rightarrow C^{rur}(d) \approx B_{cf}^{city} \quad (8)$$

For small city fraction, negligible city spreads and negative background deviations lead to Lenschow urban increments that overestimate the urban impact. For large city fractions both terms become positive and lead to Lenschow urban increments that underestimate the urban impact. In between lies the implicit city fraction size that fulfills condition (8). This is depicted for the city of Paris in Fig. 7 where condition (8) is fulfilled for a city fraction size in between the urban core and the inner city.

Unfortunately, the city fraction size associated to the Lenschow urban increment (either modelled or measured) is implicit and therefore remains unknown, unless specific model scenarios are performed to estimate it, as in this work. In addition, this implicit size is city and site dependent i.e. it will depend on the distance from the city centre ( $d$ ). For  $PM_{2.5}$ , the results indicate that Lenschow urban increments are representative of relatively small city fractions, much smaller than both their FUA and their urban core.

According to studies of several EU cities, the estimated urban impact using the Lenschow incremental approach is quite low (Querol et al., 2004). These authors report  $PM_{10}$  measured urban impacts ranging between 35% and 50% in most central Europe and Spanish cities and in the range 17–50% in Sweden and in the Netherlands. For  $PM_{2.5}$ , these increment-based urban impacts vary from 15% (Sweden) to 40%



**Fig. 6.** Comparison of the Lenschow urban increment (red) with CTM urban impacts for different city fractions. The Lenschow urban increments are estimated as average of the values reported far away from the city (but these values show a low variability as shown in Fig. 3–5). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(Spain). From the modelling side, Kiesewetter et al. (2015) for PM<sub>10</sub>, Petetin et al. (2014) and AIRPARIF (2012) for PM<sub>2.5</sub> all report increments that lead to urban impacts in Paris ranging between 30 and 35%. On the same basis, Timmermans et al. (2013) report increment-based impacts that vary between 11% in summer to 16% in winter-time, for the same city. Kiesewetter et al. (2015) report an increment-based impact around 25% for London (PM<sub>10</sub>) while Mues et al. (2013) report 46% for the city of Berlin for PM<sub>10</sub>.

Given the points discussed above, it is likely that the city fraction size implicitly considered for Paris or London in those assessments is closer to the inner city or urban core than to its FUA. The same reasoning holds for the IIASA assessment (Kiesewetter and Amann, 2014) in which PM<sub>2.5</sub> urban impacts are relatively low (below 40%), implying that most of the EU cities are net-importer of their pollution. In the latter work, the assumption made that all secondary particulate matter is of regional origin tends to reduce further the urban impacts for some cities.

### 3.3. Urban impacts for NO<sub>2</sub>

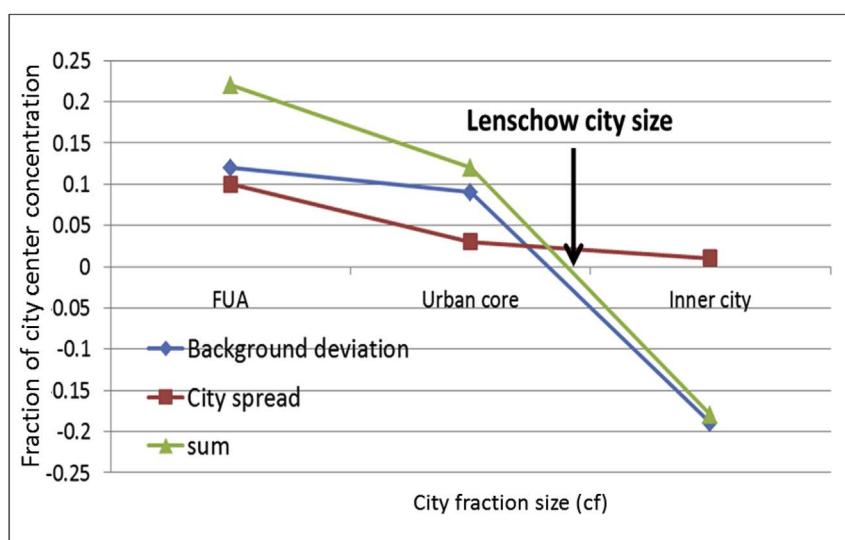
In urban environments, nitrogen oxides (NO<sub>x</sub>) are produced mostly by fossil fuel combustion. They can be emitted directly as NO<sub>2</sub> but most NO<sub>x</sub> emissions arise in the form of NO that rapidly establishes equilibrium with NO<sub>2</sub> through reaction with O<sub>3</sub>. At night, the oxidation of NO<sub>2</sub> by O<sub>3</sub> leads to the formation of NO<sub>3</sub>. The principal sink for NO<sub>x</sub> in the daytime is the formation of nitric acid (HNO<sub>3</sub>) that is removed in

about 1 week by dry and wet deposition. The resulting residence time of NO<sub>2</sub> in the atmosphere is on the order of few hours to 1 day (Wallace and Hobbs, 2006; Beirle et al., 2011). This pollutant has therefore more local characteristics than particulate matter. The two main assumptions (5) and (6) imply for NO<sub>2</sub> that:

- NO<sub>2</sub> emitted or quickly formed from NO in the city does not reach the rural location [Assumption I].
- NO<sub>2</sub> emitted by other cities or by activity sectors outside the city area (e.g. industry, shipping ...) influence the rural and city centre locations in the exact same way [Assumption II].

In the following section, we assess how those two assumptions are fulfilled as a function of the city fraction and distance from the city centre in different cities. Given the more local character of NO<sub>2</sub> we focus our analysis on the urban cores (Fig. 8).

The city spread becomes negligible at relatively short distances from the city centre (~30–40 km), but the background deviation remains significant for some cities. While the Lenschow urban increment is a very good approximation of the CTM urban impact in Paris and Berlin, it is not the case for Brussels and London, as both are characterised by significant negative background deviations. Those indicate the influence of the emissions within the commuting zone (FUA area - urban core) on the city concentration. For these two cities, a larger city fraction is needed to find a better agreement between the Lenschow urban increment and the CTM urban impact, but with the risk of further



**Fig. 7.** Background deviation and city spread components as a function of the city fraction size. Values are taken 50 km away from the city centre. The example is for the city of Paris. The sum of the two terms is used to indicate the degree of fulfilment of condition (8).

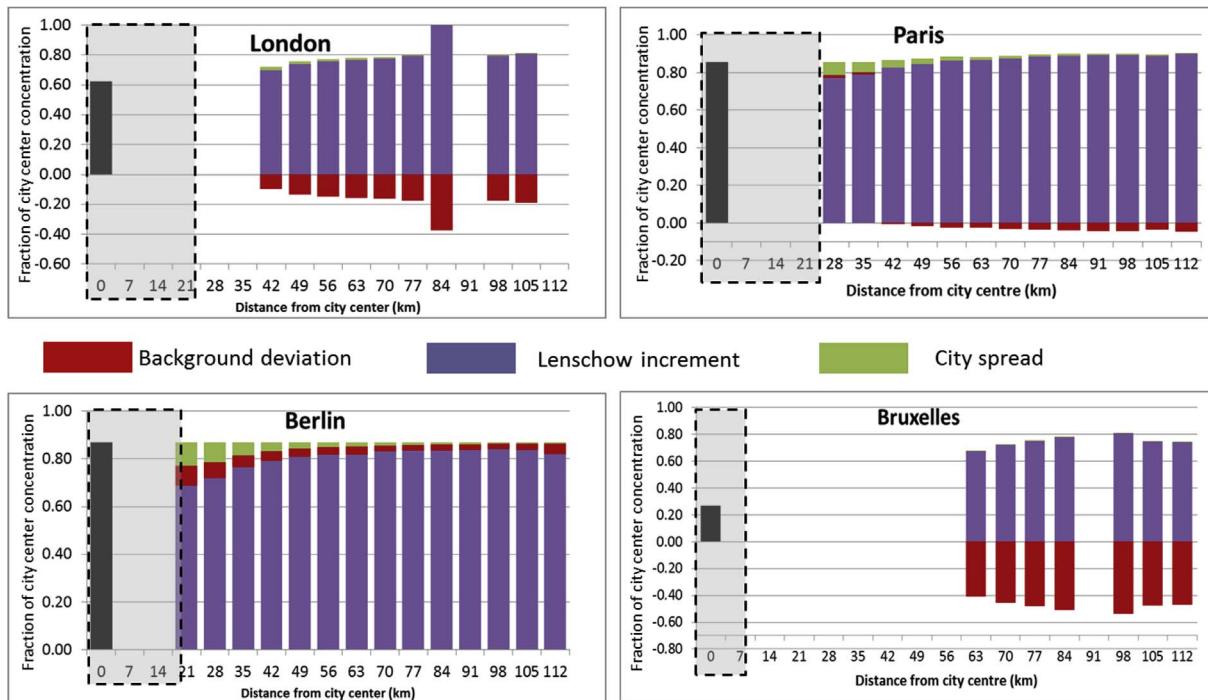


Fig. 8. Same as for Fig. 3 and 4 but for NO<sub>2</sub>. Results are calculated for the urban cores.

increasing background deviations. Fig. 9 shows that a city fraction the size of the FUA would be adequate for London, while even the FUA is not big enough for Brussels. For this city, the CTM urban impact is relatively small (around 30%) and relatively small background deviations easily bias the estimate. The fact that very few cells within the London and Brussels neighbouring region fulfil the constraint on rural emission (ratio of the rural to city NO<sub>x</sub> emissions less than 5%) illustrates the difficulty of finding appropriate location pairs to calculate the Lenschow increment. In general, the agreement is best when the difference between city centre emissions and those from the surrounding rural area around the city is large as for Paris and Berlin. Similarly to PM<sub>2.5</sub> we can identify an implicit Lenschow size for NO<sub>2</sub>, as illustrated in Fig. 9: close to FUA for London, close to the urban core for Paris and Berlin but none for Brussels (as explained above).

Similarly to PM<sub>2.5</sub>, the city fraction is a key parameter for NO<sub>2</sub> and the urban impacts vary from 35 to 100% in Paris, from 30 to 60% in Brussels, from 85 to 100% in Berlin and from 45 to 80% in London, as a function of the city fraction size considered. Although the Lenschow urban increment and the CTM urban impact sometimes agree better for NO<sub>2</sub>, the main issues reported for PM<sub>2.5</sub> remain valid. In particular, the city fraction associated to the Lenschow urban increment is unknown a-

priori and is city and distance dependent. Interpreting Lenschow urban increments as CTM urban impacts can therefore be misleading, regardless of the pollutant and city considered.

#### 3.4. Modelled vs. measured urban increments

As mentioned above, urban impacts cannot be measured. In fact, among the four terms in equation (3), only the Lenschow urban increment is measureable. For the city spread, measurements are not capable of distinguishing the city from the rural contribution within a concentration at a given location while for background deviations, zero city emissions would be required before performing measurements! As a consequence, a comparison between modelled CTM urban impacts on one side and measured Lenschow urban increments on the other would lead to erroneous interpretations as the full CTM urban impact (modelled) would be compared to only one of its components (modelled or measured). The fact that the city fraction and the distance from the city centre can be tuned to match the urban impact and the Lenschow urban increment is a proof that this comparison is ill-posed.

It remains however interesting to compare modelled and measured Lenschow urban increments. This comparison is seldom performed

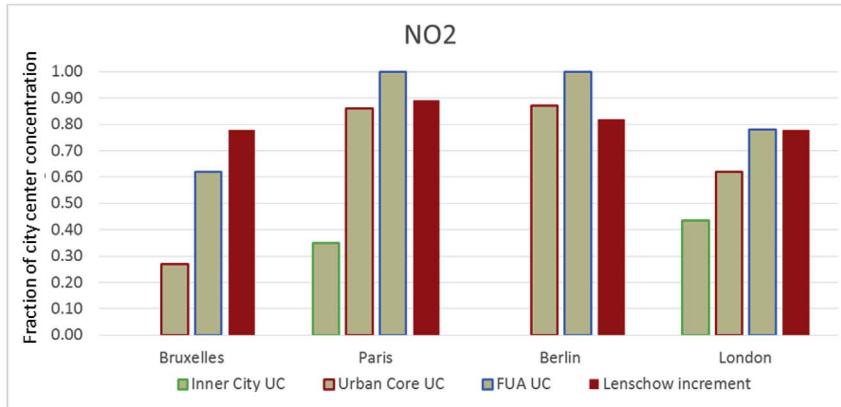
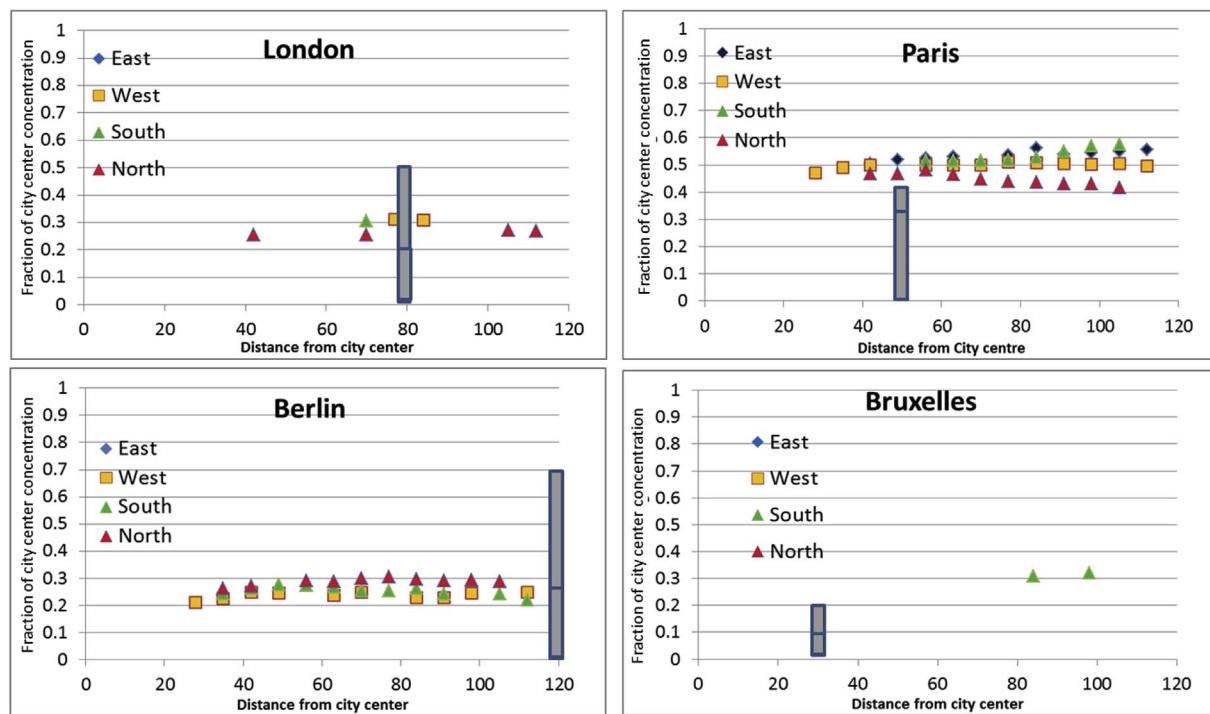


Fig. 9. Comparison of the urban impact (red) with Lenschow increments obtained with different city fraction sizes (areas over which emissions are reduced). The Lenschow increments are estimated as an average of the values reported far away from the city (but these values show a low variability as shown in Fig. 8. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 10.** Variation of the modelled Lenschow urban increments for  $\text{PM}_{2.5}$  in terms of distance ( $d$ ) from the city centre. The increments are estimated along the four main geographical directions [East (E), West (W), North (N) and South (S)]. The grey bars indicate the values of the measured increments obtained from the available monitoring AIRBASE stations (see Annex for details). If more than one station is available, the minimum and maximum values are indicated via the blue dividing line. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(Mues et al., 2013; Timmermans et al., 2013; Petetin et al., 2014), as the focus is generally on comparing modelled and measured time series at selected locations, provides information on the CTM ability to correctly reproduce the measured spatial gradients between the city and its rural environment. Using this approach, Timmermans et al. (2013) highlighted the impact of the underlying emission inventory on the calculation of urban increments for the city of Paris. Petetin et al. (2014) extended this analysis to chemical composition (e.g. nitrate, sulphate ...) and to shorter periods, a temporal scale at which physical and chemical processes can be identified more easily. In their comparison of a CTM with measurements in Paris, they showed that errors in ammonium nitrate were compensating for by the other compounds. It is interesting to note that this analysis can directly be performed from the base-case simulations and therefore does not require intensive CPU resources.

Based on pairs of locations, the comparison of modelled and measured Lenschow urban increments is very sensitive to the choice of locations. This is why the Lenschow urban increments are calculated and shown along each geographical direction (Figs. 10 and 11). For each city, the available urban background and rural background monitoring stations (see details in the annex) are used to calculate a minimum and maximum Lenschow urban increment for each rural station, reported as function of their distance from the city centre ( $d$ ). Measurement data consist of yearly average values for the year 2010, with the exception of Paris for which  $\text{PM}_{2.5}$  measurements are those of 2013 due to their unavailability before that date (see Annex for details). The modelled Lenschow increment (modelled spatial concentration gradient) is directly extracted from the CHIMERE base case simulation.

The comparison highlights the difficulties in the selection of a representative pair of stations and their use to subsequently evaluate model increments. In the case of Brussels, the two selected urban stations report values that differ sufficiently to generate a factor 2 in the estimate of the Lenschow urban increments (range from 10 to 20%), regardless of the concentration level measured at the rural station. The same is seen in London where the measured Lenschow urban

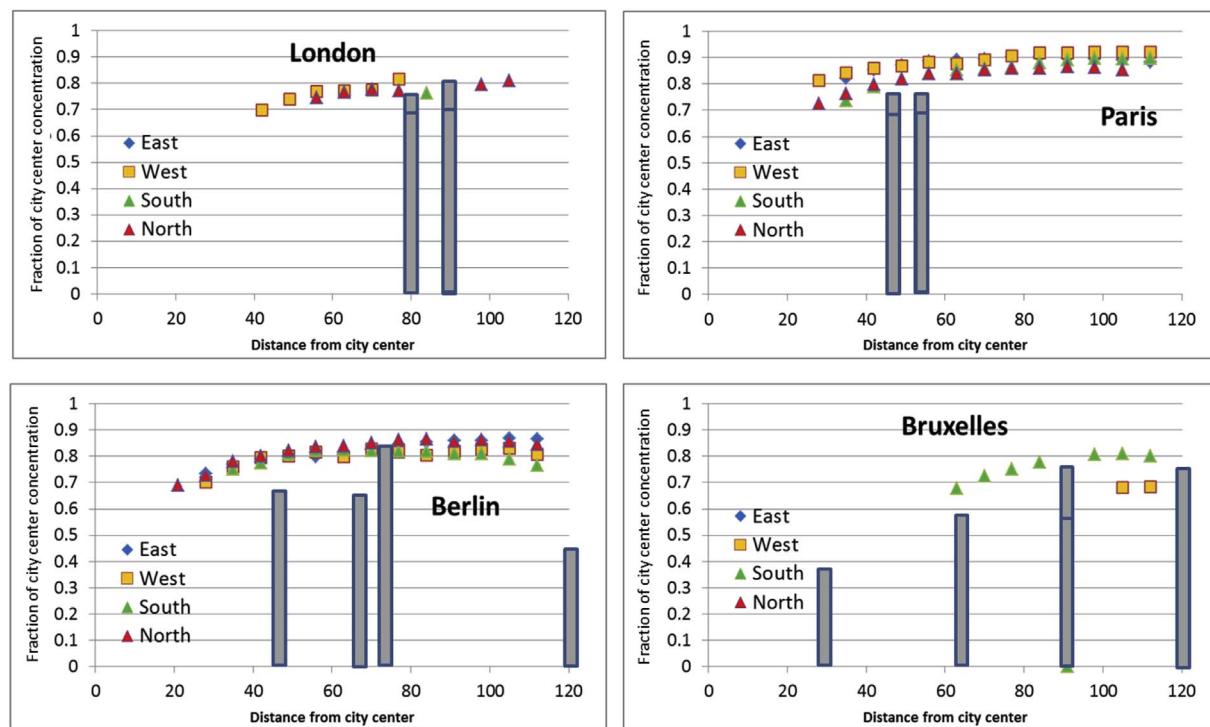
increments range from 20 to 50%, depending on the selected urban background station. While urban values are averaged in some works (Kiesewetter and Amann, 2014; Mues et al., 2013) to obtain a single increment value, here we keep the individual values to highlight the sensitivity of the increment to the urban variability. The same argument holds for the rural background stations, for which the sensitivity of the Lenschow increment is clearly highlighted by its variability in terms of distance (Figs. 10 and 11). A good example is Berlin where the available rural stations located beyond 120 km lead to Lenschow increments varying between 25 and 70%. Although the  $\text{NO}_2$  results are more consistent, similar issues remain, mostly because of the difficulty in removing all background deviation.

The fact that Lenschow urban increments are very sensitive to distance and to the choice of the rural background station supports the CTM model analysis above and is an additional fact pointing to the difficulty of using these increments as proxies for estimating urban impacts.

#### 4. Conclusions

Recent air quality assessments (Kiesewetter et al., 2015; Kiesewetter and Amann, 2014), performed at the European scale, now provide information on source apportionment and in particular on the fraction of the air pollution attributed to different geographical entities: urban, country or transboundary. The main purpose of providing this information is to improve air quality policy governance by supporting authorities in choosing the most efficient actions at the appropriate administrative level and scale of decision. The question of estimating the impact of cities to air quality is therefore not only theoretical as it is critical to the identification of effective strategy options related to air quality, in particular the choice between local actions focusing on the urban scale and actions requiring national/international efforts.

In this context, it is important to compare the methodologies that are available to estimate the urban impact on air pollution. As the urban impact is not measurable, different approaches have been proposed to



**Fig. 11.** Variation of the modelled Lenschow urban increments for NO<sub>2</sub> in terms of the distance (d) from the city centre. The increments are estimated along the four main geographical directions [East (E), West (W), North (N) and South (S)]. The grey and blue bars indicate the minimum and maximum values of the measured increments obtained from the available AIRBASE monitoring stations (see Annex for details). If more than one station is available, the minimum and maximum values are indicated via the blue dividing line. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

circumvent this problem. In this work, we compared two main approaches: one based on modelling (CTM urban impact) and the other based on spatial increments (Lenschow urban increment), either measured or modelled. We showed that the urban impact is the sum of three terms: the Lenschow urban increment, the city spread and the background deviation, with the Lenschow urban increment being representative of the urban impact only when the two latter terms are negligible. This latter condition was expressed in the form of two main assumptions that need to be fulfilled. The first assumption implies that the rural background location is not influenced by the city emissions whereas the second requires that background concentration levels, obtained when the city emissions are switched off, are equal at both locations.

Current knowledge in atmospheric chemistry and dispersion processes indicates that cities influence air quality in their neighbouring rural areas. This is especially true for particulate matter, characterised by relatively long residence times in the lower troposphere (up to several days), and consequently travel long distances (of the order of a thousand kilometres). The first assumption (negligible city spread), which implies finding a rural background location where no city impact is felt, is therefore likely to be violated to some extent. The second assumption, i.e. on the homogeneity of the background levels is also questionable in many European cities due to the presence of many other potential emission sources (other cities, industry ...) in vicinity of the urban area. While these assumptions are generally recognised, the extent of their fulfilment is never quantified. This is due in particular to the impossibility of performing this assessment via measurements. In this work, we used a modelling approach (SHERPA) to quantify all three components (Lenschow urban increment, city spread and background deviation) in some European cities as a function of the city fraction. The main aim was to identify under which conditions the incremental approach leads to a robust estimate of the urban impact. The analysis was performed both for PM<sub>2.5</sub> and NO<sub>2</sub>.

Model results indicate that for PM<sub>2.5</sub>, these two assumptions are far

from being met for large (FUA) or medium (urban core) city fractions. As a consequence, Lenschow urban increments largely underestimate the urban impacts of the broadest definitions of cities by up to a factor 2. Although results are in better agreement for NO<sub>2</sub>, i.e. the incremental approach leads to more accurate estimates of the urban impact, similar issues remain, mostly because of the difficulty in avoiding any background deviation, that strongly depends on the homogeneity of the land-use around the city. Satisfying these assumptions is city, distance and pollutant specific, consequently no generalisation can be made, although in our analysis the Lenschow urban increment can be a valid estimator of the urban impact for city fractions between the inner city and the urban core.

For both PM<sub>2.5</sub> and NO<sub>2</sub>, the city fraction is the key factor influencing the urban impact, with factors of 2 or 3 differences predicted when moving from small (inner city) to large (FUA) city fractions. Given the latter point and the fact that (1) the fraction of the real city, represented by the Lenschow approach, is unknown, as it is city, distance and pollutant specific and (2) a factor of 2 difference can easily exist for two sets of locations pairs for the same city fraction, using the Lenschow incremental approach to estimate the urban impact on air pollution can produce quite uncertain results. As illustrated in this work, this can in turn lead to sub-optimal choices in the frame of urban air quality planning.

Because both assumptions can only be assessed via modelling, the quality of the model (and of its associated inputs, e.g. meteorology, emissions ...) is crucial. In this respect, the ability of a model to reproduce spatial concentration gradients between the city and its rural surroundings is a feature that can be tested by comparing measured and modelled Lenschow increments. Some examples of such comparisons have been presented for the cities considered in this analysis and should be further extended, although the selection of the station represents a critical step. It is however not sufficient to ensure that we accurately model urban impacts as this will also depend on the ability of the CTM to reproduce specific emission scenarios (e.g. Clappier et al., 2017;

Thunis et al., 2015). In this respect, CTMs inter-comparison exercises probably represent the only practical approach to improve our confidence in CTM responses to emission changes. It is worthwhile mentioning that past inter-comparison exercises at country (Bessagnet et al., 2016; Thunis et al., 2010), regional (APRLR, 2011) or city scale (Cuvelier et al., 2007; Thunis et al., 2007) all highlighted the fact that models are more robust for scenarios (deltas) than for predicting base case concentrations, a comforting fact regarding the calculation of urban impacts.

The objective of this work was to highlight the challenges related to the estimation of urban impacts on air quality. This analysis was limited to background levels, to yearly averages and to total mass in the case of PM<sub>2.5</sub>. However, the same analysis could be extended to shorter

temporal averages (daily, hourly, seasonal ...) to different species (e.g. sulphate, nitrate ...) and/or sectors (transport, industry ...) for a better understanding of each process. It is also interesting to note that a similar approach could be used to assess the impact of street emissions to the urban background in cities (street impact vs. street increment), especially for NO<sub>2</sub>. Different underlying models (better spatial resolution) would however be required then to perform this task.

### Acknowledgments

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### Annex

The available measurements used to calculate the urban increments (*LUI*) for the 4 cities analyzed in this work are listed in the tables below for PM<sub>2.5</sub> and NO<sub>2</sub>. The minimum increment for a given city is calculated as the difference between the minimum of the concentrations reported at the urban background locations on one hand and the maximum of the concentrations reported at the rural locations, on the other. The maximum increment for a given city is calculated as the difference between the maximum of the concentrations reported at the urban background locations on one hand and the minimum of the concentrations reported at the rural locations, on the other.

$$LUI_{max} = \max[C_{city}] - \min[C_{rur}]$$

$$LUI_{min} = \min[C_{city}] - \max[C_{rur}]$$

Table 2

NO<sub>2</sub> measurement stations used for the calculation of the urban increments in Figs. 10 and 11 (RB – Rural background, UB – Urban background, SB – Suburban background)

Station	Longitude	Latitude	Station type	Distance	Concentration	Lenschow	
						Min	Max
Kensington	-0.21	51.51	UB		36.83		
Westminster	-0.13	51.49	UB		51.98		
Harwell	-1.33	51.57	RB	80	11.94	68%	77%
Wicken Fen	0.29	52.30	RB	92	11.09	70%	79%
6eme Arond	2.34	48.85	UB		35.8		
1er Arond	2.35	48.86	UB		44.38		
7eme Arond	2.29	48.86	UB		44.38		
Sonchamp	1.88	48.58	RB	46	11	69%	75%
Fontainebleau	2.65	48.35	RB	55	11.13	69%	75%
Parlement Europ.	4.37	50.84	UB		36.58		
St Pieters Leeuw	4.22	50.77	RB	10	21.58	41%	41%
Corroy le Grand	4.67	50.66	RB	30	23	37%	37%
Andenne	4.99	50.50	RB	65	16.11	56%	56%
Somme Leuze	5.23	50.28	RB	91	15.9	57%	57%
Viroinval	4.59	50.10	RB	87	9	75%	75%
Sainte Ode	5.59	50.03	RB	123	10	73%	73%
B. Mitte BruckenStr.	13.42	52.51	UB		27.94		
B. Neuklin NansenStr.	13.43	52.49	UB		28.39		
B. wedding	13.35	52.54	UB		28		
B. Grunewald	13.23	52.47	RB	128	15.38	45%	46%
B Friedrichshagen	13.65	52.45	RB	16	13.49	52%	52%
Lutte	12.56	2.19	RB	66	9.66	65%	66%
Buckow	14.02	52.56	RB	46	9.14	67%	68%
Neuglobsow	13.03	53.14	RB	75	5.28	81%	81%
Spreewald	14.06	51.90	RB	75	7.88	72%	72%

Table 3

Same as Table 2 but for PM<sub>2.5</sub>

Station	Longitude	Latitude	Station type	Distance	Concentration	Lenschow	
						Min	Max
Kensington	−0.21	51.51	UB		10.84		
Westminster	−0.13	51.49	UB		12.64		
Bloomsbury	−0.13	51.52	UB		16.40		
Harwell	−1.33	51.57	RB	80	8.64	20%	47%
Vitry sur Seine	2.38	48.78	UB		17.32		
Bobigny	2.45	48.90	UB		16.79		
Genevilliers	2.29	48.93	UB		16.42		
Paris 4eme	2.35	48.86	UB		18.65		
Bois Herpin	2.24	48.36	RB	54	11.14	32%	40%
Uccle	4.36	50.80	SB		18.45		
Berchem	4.29	50.86	SB		16.98		
Corroy le Grand	4.67	50.66	RB	30	15.21	10%	18%
B. Mitte BruckenStr.	13.42	52.51	UB		20.38		
B. Neuklin NansenStr.	13.43	52.49	UB		21.48		
B. wedding	13.35	52.54	UB		19.84		
Wurmberg	10.61	51.76	RB	208	6.34	69%	70%
Friedrichsbrunn	11.04	51.66	RB	186	14.96	27%	30%
Collmberg	13.01	51.30	RB	136	13.69	33%	36%
Wendland	11.17	52.96	RB	156	12.99	36%	40%
Waldhof	10.76	52.80	RB	180	15.07	26%	30%

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