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## More mileage in reducing urban air pollution from road traffic

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### ABSTRACT

Road traffic emissions are considered a major contributor to urban air pollution, but clean air actions have led to a huge reduction in emissions per vehicle. This raises a pressing question on the potential to further reduce road traffic emissions to improve air quality. Here, we analysed ~11 million real-world data to estimate the contribution of road traffic to roadside and urban concentrations for several major cities. Our results confirm that road traffic remains a dominant source of nitrogen dioxide and a significant source of primary coarse particulate matter in the European cities. However, it now represents a relatively small component of overall PM<sub>2.5</sub> at urban background locations in cities with strong controls on traffic emissions (including European cities and Beijing) and many roadside sites will exceed the WHO guideline (10 µg m<sup>-3</sup> annual mean) even when this source is eliminated. This suggests that further controls on traffic emissions, including the transition to a battery-electric fleet, are needed to reduce NO<sub>2</sub> concentrations, but this will have limited benefit to reduce the concentration of fine particles, except in countries where the use of diesel particle filters is not mandatory. There are substantial differences between cities and the optimal solution will differ from one to another.

### 1. Introduction

Emissions from road traffic make an appreciable contribution to overall air pollution (Amato et al., 2014; Grange et al., 2017; Pant and Harrison, 2013; Yin et al., 2015), although the common assumption that traffic is the dominant factor in urban pollution may not always be correct. Outdoor air pollution is a major cause of premature deaths (Chen et al., 2017; Zhang et al., 2019; WHO, 2013; Kheirbek et al., 2016) and is ranked fifth in the causal factors for non-communicable disease mortality (Cohen et al., 2017). Particulate matter pollution (PM) alone caused 4.2 million premature deaths in 2015 worldwide (Cohen et al., 2017), largely in developing countries such as China and India. Air quality in western countries has improved enormously in the past decades and the reductions of road vehicle emissions have been a major contributor to this trend. However, the question which now has to be faced is what future contribution can be expected from further reductions in emissions from the road vehicle fleet. On the one hand, many scientists continue to argue that traffic is still a major source of air

pollution (Amato et al., 2014; Chen et al., 2018; Grange et al., 2017; Li et al., 2017; Pant and Harrison, 2013; Yin et al., 2015), while remote sensing data point to significant emissions reductions for some pollutants (Carslaw et al., 2019) and motor industry scientists point to the large reductions in emissions per vehicle over the past 30–40 years and question how much further reduction in emissions is warranted (Winkler et al., 2018).

Road traffic contributes to air pollution through a range of mechanisms (Pant and Harrison, 2013; Amato et al., 2014), including:

- Emissions of particulate matter and gases in engine exhaust.
- Secondary air pollutants formed from primary emissions.
- Non-exhaust emissions including wear of the brakes, tyres and road surface and resuspended dust. Unlike exhaust emissions, non-exhaust emissions are not subject to legislative control currently.

Here, the concentration impacts of primary emissions of pollutants upon air quality in the immediate vicinity and more widely in six cities

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(London, Paris, Berlin, Beijing, Istanbul and Hong Kong) were evaluated based on measured data from national network monitoring sites downloaded from national archives. Since meteorological conditions change year by year, leading to variations in the concentration of air pollutants even when the emissions remain the same, we firstly applied a machine-learning based Random Forest algorithm to decouple the effect of meteorological conditions to obtain the weather normalized air pollutant concentrations (Vu et al., 2019; Grange et al., 2018). This approach has the advantage over air quality modelling that it does not depend upon use of emission factors, which are often uncertain (Li et al., 2017; Franco et al., 2013). The increments due to traffic and urban emissions were then calculated from the de-weathered air quality data.

- (a) *Roadside increment: by subtracting air pollutant levels at urban background site(s) from those made at a trafficked site.* This is a measure of the impact of traffic on an urban road above the background urban air quality. This is not the entire contribution of road traffic to the urban air pollution levels, but represents a major component which is directly attributable to road traffic.
- (b) *Urban increment: by subtracting air pollutant levels at rural background site(s) from those made at urban background site(s).* This provides an estimate of the impact of all local urban sources, including traffic, upon air quality. Similar to roadside increment, the higher the value, the larger the contribution of urban air pollutant emissions to air quality. For some pollutants in some cities the urban increment is mainly due to traffic, while for others traffic is just one contributor amongst several.

Such methods have previously been used successfully (Jones et al., 2005; Hilker et al., 2019) and show the immediate local benefits which would accrue from a requirement for zero emissions on a highway (roadside increment). Since air pollution climates vary significantly around the world, we analysed datasets from both developed and developing countries.

## 2. Materials and methods

### 2.1. Air sampling sites

Air quality monitoring sites were carefully chosen to cover as many types (roadside, urban background, suburban and regional background) as possible.

### 2.2. Beijing

In the case of Beijing, data have been taken from a total of 35 national and local monitoring stations, including 12 urban background sites, 11 suburban sites, 7 remote/rural sites and 5 traffic sites. The five traffic monitoring sites can be divided into three groups; three stations are on inner urban roads with a large flow of light duty gasoline vehicles and two are on major arterial highways in the area of the third and fourth ring roads with a high volume of diesel trucks.

### 2.3. London

In the case of London, spatial variation in background air pollution is much less; the annual mean PM<sub>2.5</sub> at background sites (n = 7) in London in 2018 ranged from 9 to 11 µg m<sup>-3</sup>. The London North Kensington station has been characterised in detail by Bigi and Harrison (2010) and has been used widely as indicative of background air quality in central London. Roadside data are taken from the Marylebone Road site which is on a major arterial highway in central London, with six lanes carrying 80–90,000 vehicles a day within a street canyon. This represents one of the most polluted roadside sites in London. Background data for the UK are taken from the rural Harwell and Chilbolton sites which have been selected because of their freedom from local influences and

representativeness of southern England.

### 2.4. Paris

The roadside site in Paris (Périphérique Auteuil) is at roadside on the Paris ring road which, at this point, is an eight lane highway carrying around 240,000 vehicles per day. It is compared with a background site at Vitry-Sur-Seine which lies within a residential area in the south of Paris, about two kilometres away from the ring road. The rural site data was collected at two monitoring stations:

- (a) Southern rural area - Herpin wood which is located in Bois-herpin, in the south of Paris (~60 km); data for PM<sub>10</sub>, PM<sub>2.5</sub> and O<sub>3</sub>
- (b) Zone rurale Sud-Est - Forêt de Fontainebleau, which is located in Maison Forestière du Clos du Roi, Fontainebleau in the south-east of Paris (~62 km); data for PM<sub>10</sub>, PM<sub>2.5</sub> and NO<sub>2</sub>. The final datasets of PM<sub>10</sub> and PM<sub>2.5</sub> were estimated as the averaged values from datasets obtained from two above stations. Data were downloaded from: <https://www.airparif.asso.fr/en/indices/resultats-jour-citeair#jour>.

### 2.5. Hong Kong

The monitoring sites are classified into traffic, urban background and rural sites by the Environmental Protection Department, Government of Hong Kong.

For roadside sites, the values were calculated as a median value from three road monitoring stations: Causeway Bay, Central and Mong Kok which are urban roadside in a mixed residential/commercial area with heavy traffic and surrounded by many tall buildings.

The urban background site values were calculated as a median value from five urban monitoring stations: Sha Tin, Tai Po, Tung Chung, Yuen Long & Tuen Mun which are located in mainly residential areas in New Town. The rural site is Tap Mun, located at Tap Mun Police Post, and is classified as rural background.

Link to for the monitoring data and site information: <https://www.aqhi.gov.hk/en.html>.

The monitoring sites are classified into traffic, urban background and rural sites by the Environmental Protection Department, Government of Hong Kong.

### 2.6. Istanbul

For the traffic sites, the concentrations were calculated as a median value from five roadside monitoring stations: Aksaray, Catladikapi, Selimiye, Umraniye and Besiktas. For the urban sites the concentrations were calculated as a median value from three urban monitoring stations: Umraniye, Maslak, Avcilar. The rural site concentrations were calculated as a median value from four rural monitoring stations: Arnavutkoy, Sile, Buyukada, and Kumkoy.

For more information for the site classification (in Turkish): <https://mthmm.csb.gov.tr/hava-kalitesi-olcum-istasyonlarimiz-i-85693>, and for the monitoring data: <https://www.havaizleme.gov.tr/Services/AirQuality/Default.ltr.aspx>

### 2.7. Berlin

The roadside site (station code: MC174/DEBE064) is located on a major highway (Franfurter Allee; 52°30' 50" N; 13°28' 11" E) with high traffic flow in a densely populated city centre area. The urban background site (station code: MC042/DEBE064) was located in an area of high population density and low traffic flow (52°29' 21.98" N; 13°28' 51.08" E). As there was no rural station available, data were taken from a suburban station (station code: MC077/DEBE051) located in Pankow,

Hobrechtsfelder Chaussee, Waldschiele (52°38' 39" N; 13°28' 59"E) in a hospital area with low traffic and nearby forest.

## 2.8. Random Forest method

Weather conditions change year by year, causing variations in the concentration of air pollutants even when the emissions do not change. Here, we applied a machine-learning based Random Forest algorithm to decouple the effects of meteorological conditions to obtain the weather-normalized air pollutant concentrations. The model was found to predict very well concentration of pollutants in testing data sets ( $r^2$  greater than 0.8). Details of the method used to remove variations caused by weather from air pollution time series datasets are given by Vu et al. (2019). Meteorological data from the last 30 years were used for normalisation. All concentrations reported here refer to weather-normalized unless mentioned otherwise.

## 3. Results

We firstly examined the urban and roadside increments in Beijing, which has an extensive air quality network with a total of 35 sites (5 roadside, 23 urban background, and 7 remote/rural sites). Fig. 1 shows the time series of roadside and urban increments in weather-normalized weekly concentrations of six criteria pollutants at roadside, urban background and rural sites in Beijing from 2014 to 2018. Roadside and urban  $PM_{2.5}$  increments significantly decreased from 8.5 to 4.2  $\mu g m^{-3}$  and 8.6 to 1.8  $\mu g m^{-3}$  respectively from Jul 2014-Jun 2015 to Jul 2017-Jun 2018 (Fig. 1). On the other hand, there was almost no change in the  $NO_2$  urban increment during this period. The  $SO_2$  roadside increment decreased from 3.2 to 2.2  $\mu g m^{-3}$  but the urban increment reduced more dramatically. CO roadside increments decreased by more than 50%. Both roadside and urban increments for Beijing are a small proportion of

the total pollutant concentration, except for  $NO_2$ , for which roadside concentrations far exceed the rural background (Figure S1; Table 1). Ozone, as expected, shows a roadside decrement in concentration due to titration by NO from vehicle emissions. The air pollution in the Beijing area is a regional phenomenon with a large contribution from secondary sources to particulate matter (Huang et al., 2014; Liu et al., 2019) but even the primary pollutants (e.g. CO) with the exception of  $NO_2$  show remarkable similarity between site types (Figure S1). As shown in Vu et al. (2019), there is an increasing trend in maximum (summer) deweathered  $O_3$  in Beijing, which is consistent with the trend observed by Li et al. (2019).

The roadside and urban increments in the six cities for the period of 2016–2018 are shown in Table 1 and Figs. 2 and 3.  $PM_{2.5}$  roadside traffic increments in the six cities from 2016 to 2018 are broadly similar, ranging from  $\sim 2 \mu g m^{-3}$  in Berlin and Istanbul to  $\sim 5 \mu g m^{-3}$  in Paris, Beijing, London and Hong Kong (Table 1a). However, the percentage increase from the urban background levels by traffic ranges from 7.5% (Beijing) to nearly 50% (London). This is reflected in Fig. 2, which shows the large roadside increment in London and Paris but small increment in Beijing and Berlin. Except for London, the urban increment above rural concentrations is of similar magnitude to the roadside increment above urban background (Fig. 2). Evidence is also available from the United States Near-Road Monitoring Network. Data from 31 sites (2014) and 47 near-road sites (2015) were analysed in relation to sites representative of the local background. Near-road increments were calculated for  $PM_{2.5}$  using several methods to estimate the most appropriate background concentration (DeWinter et al., 2018; Seagram et al., 2019; Brown et al., 2019). The analysis for 2014 and 2015 data revealed an average roadside increment for  $PM_{2.5}$  of  $1.2 \mu g m^{-3}$  (range  $-1.2 \mu g m^{-3}$  to  $3.1 \mu g m^{-3}$ ) across 26 cities (DeWinter et al., 2018). Analysis of data for 2016 showed a mean roadside increment of from  $0.6 \mu g m^{-3}$  to  $1.1 \mu g m^{-3}$  depending on the method of estimating the background

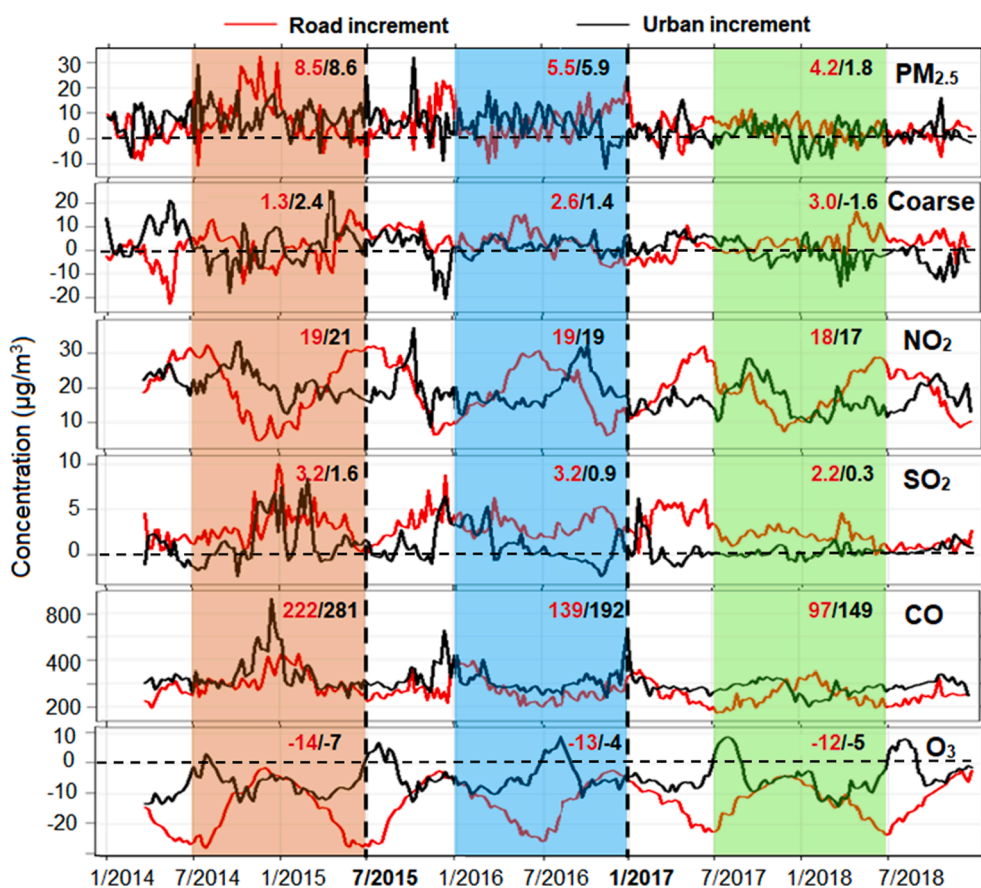


Fig. 1. Roadside and urban increment in Beijing during 2014–2018. The three shaded areas represent three periods for comparison of pollutant concentrations (1) July 2014 – June 2015: before the implementation of the China V standard for all vehicles (including freight trucks and long-distance coaches, Jun 2015), (2) Jan–Dec 2016; after the implementation of China V for all vehicles; (3) July 2017 – Jun 2018 after the implementation of the China 6/VI fuel standards (Jan 2017) + traffic restriction of light-duty gasoline passenger cars of China 1/2 (Feb 2017). The numbers shown in the figures represent the roadside and urban increments (red and black) during the shaded periods. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

**Table 1**

Average (a) roadside and (b) urban increment ( $\mu\text{g m}^{-3}$ ) and percentage increase above (a) urban background, and (b) rural concentration of air pollutants in various cities during 2016–2018.

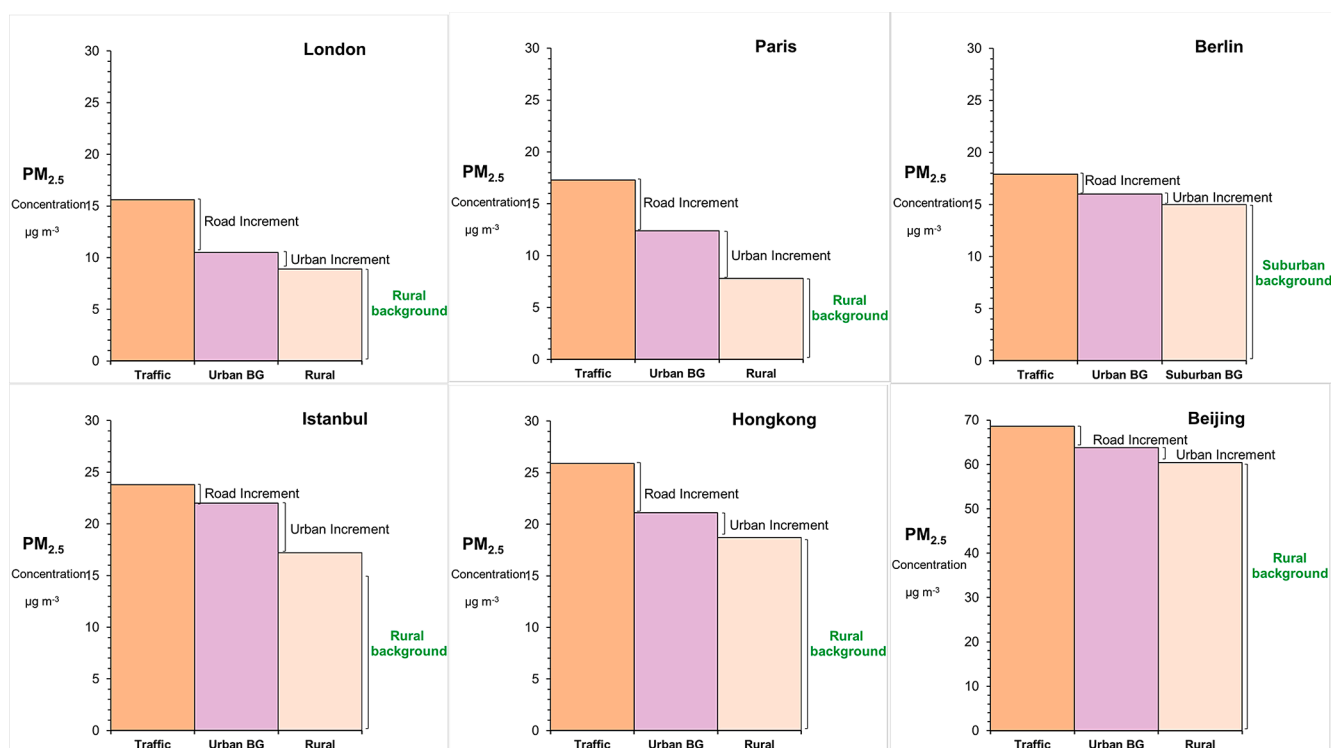
a	PM <sub>2.5</sub>		PM <sub>10</sub>		PM <sub>2.5-10</sub>		NO <sub>2</sub>		NO <sub>x</sub>	
	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase
London	5.1	48.6	8.1	48.5	3.0	48.4	54.9	170.0	230.3	460.6
Paris	4.9	39.5	15.0	71.1	10.1	116.1	58.6	192.1	n.a	n.a
Berlin	1.9	11.9	4.3	18.6	2.4	33.8	14.0	53.8	46.6	129.8
Beijing	4.8	7.5	7.2	7.2	2.3	6.4	18.7	37.9	n.a	n.a
Hong Kong	4.8	22.7	3.0	8.8	-1.8	-13.8	43.2	112.5	127.2	219.7
Istanbul	1.8	8.2	11.2	32.0	9.4	71.8	38.3	116.8	n.a	n.a

b	PM <sub>2.5</sub>		PM <sub>10</sub>		PM <sub>2.5-10</sub>		NO <sub>2</sub>		NO <sub>x</sub>	
	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase
London	1.6	18.0	2.3	16.0	0.7	12.7	20.4	171.4	35.0	233.3
Paris	4.6	59.0	7.4	54.0	2.7	45.0	23.3	323.6	n.a	n.a
Berlin	1.0*	6.7	4.0	20.9	2.1*	42.0	11.3	76.9	17.7	97.3
Beijing	3.4	5.6	2.9	3.0	-0.5	-1.4	17.4	54.5	n.a	n.a
Hong Kong	2.4	12.8	2.0	6.3	-0.4	-3.0	28.3	280.2	45.4	363.2
Istanbul	4.8	27.9	9.0	34.6	4.2	47.2	16.8	105.0	n.a	n.a

Note: \*data is only available for 2016.

The statistics are based on monthly averaged weather-normalised data, except data for Hong Kong and Istanbul which are not de-weathered; n.a. = not available; percentage increase is calculated as the roadside increment / urban background concentration \* 100 or urban increment/rural background concentration \* 100.



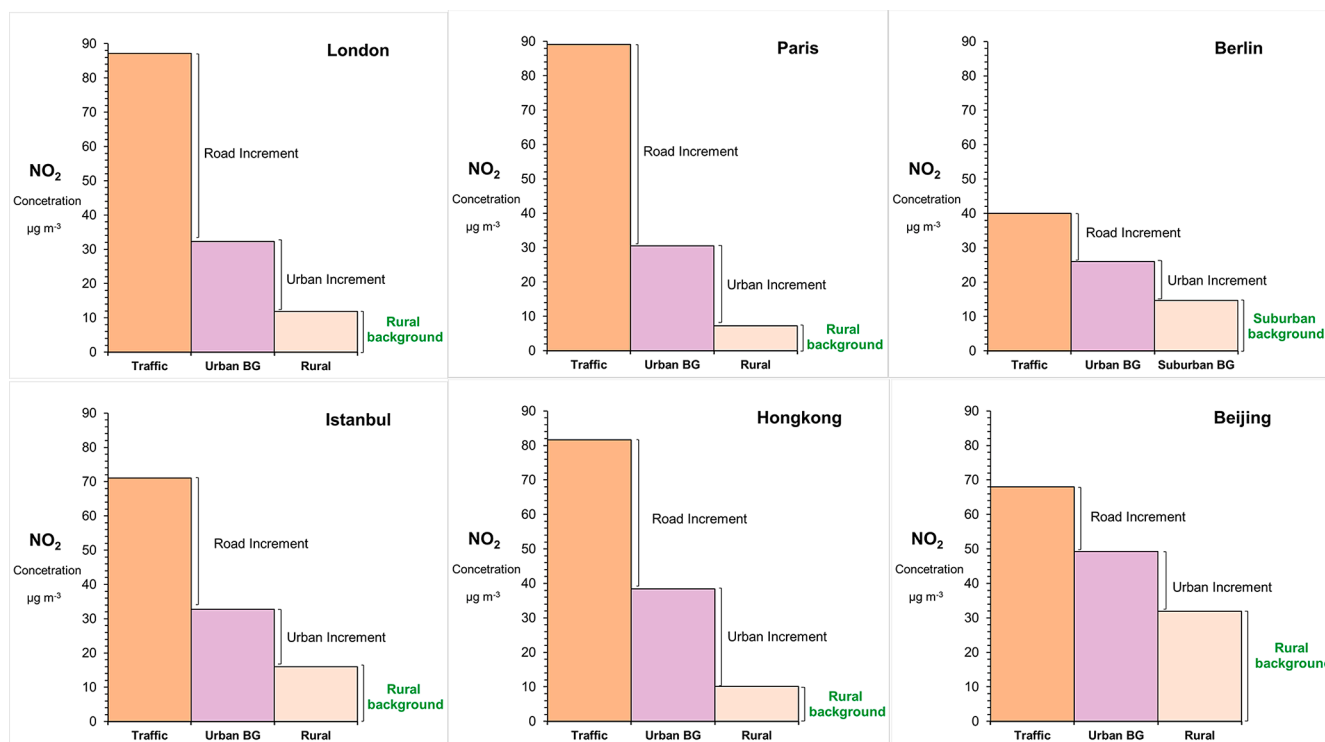
**Fig. 2.** Average roadside traffic and urban increments of PM<sub>2.5</sub> over three years (2016–2018) for the six cities. All the data are de-weathered for London, Beijing, Paris and Berlin. PM<sub>2.5</sub> road increment ranges from 1.9  $\mu\text{g m}^{-3}$  in Berlin to 5.1  $\mu\text{g m}^{-3}$  in London and urban increment from 1.0  $\mu\text{g m}^{-3}$  in Berlin to 4.6  $\mu\text{g m}^{-3}$  in Paris. It is noted that the road increment in London, Paris and Beijing are almost the same. However, the percentage road and urban increases (the roadside increment/urban background concentration \* 100 or urban increment/rural background concentration \* 100) are significantly different, from 7.5% in Beijing to 48.6% in London, and 5.6% in Beijing to 59.0% in Paris, respectively. PM<sub>2.5</sub> concentration in Beijing was much higher than those in other cities.

(Seagram et al., 2019).

The increment in coarse particles (PM<sub>2.5-10</sub>) ranges from 2.3  $\mu\text{g m}^{-3}$  in Beijing to 10.1  $\mu\text{g m}^{-3}$  in Paris from 2016 to 2018 (with Hong Kong as an exception). Similar to that of PM<sub>2.5</sub>, the percentage increase from the urban background levels by traffic is highly variable in different cities, ranging from 6.4% in Beijing to 116% in Paris (Table 1a). The roadside

PM<sub>10</sub> increment is rather small in Beijing and Berlin but quite high in London and Paris (Figure S2).

Roadside increments of NO<sub>2</sub> and NO<sub>x</sub> show a very different picture to those of the particles. The increment in NO<sub>2</sub> ranges from 14  $\mu\text{g m}^{-3}$  in Berlin to 58.6  $\mu\text{g m}^{-3}$  in Paris during 2016–2018. The NO<sub>x</sub> roadside increment is huge, at 46.6, 127.2 and 230.3  $\mu\text{g m}^{-3}$  in Berlin, Hong Kong



**Fig. 3.** Roadside traffic and urban increments of nitrogen dioxide for the six cities, average of 2016–2018. All the data are de-weathered for London, Beijing, Paris and Berlin. NO<sub>2</sub> road increment ranges from 14 µg m<sup>-3</sup> in Berlin to 58.6 µg m<sup>-3</sup> in Paris and urban increment from 11.3 µg m<sup>-3</sup> in Berlin to 23.3 µg m<sup>-3</sup> in Paris. It is noted that road increment in London and Paris is similar whereas that in Beijing and Berlin is similar. However, the percentage road and urban increases are significantly different, from 37.9% in Beijing to 192.1% in Paris, and 54.5% in Beijing to 323.6% in Paris, respectively.

and London, respectively. The percentage NO<sub>2</sub> increase from the urban background levels by traffic ranges from 38% in Beijing to 170% in London (Table 1a; Fig. 3).

Figure S3 further shows a decreasing trend of deweathered PM<sub>2.5</sub> roadside increment from 12.5 µg m<sup>-3</sup> for Paris and 9 µg m<sup>-3</sup> for London in 2009–2012 to ~5 µg m<sup>-3</sup> in 2016–2018. This is attributed to the introduction of the Euro 5 (2011) and Euro VI (2013) standards which have reduced exhaust particulate matter emissions from light and heavy duty vehicles respectively. In Beijing, the de-weathered PM<sub>2.5</sub> roadside increment in the winter months is very high in 2014 but reduced by four fold in 2019 (Figure S3). This is likely related to the implementation of China V for all vehicles in Jun 2015 and China 6/VI fuel standards in Jan 2017, plus traffic restrictions on light-duty gasoline passenger cars of China 1/2 (Feb 2017) (Fig. 1). Figure S4 shows that the increment in de-weathered coarse particles (PM<sub>2.5-10</sub>) has remained fairly constant in Paris from 2009 to 2018 at around 10 µg m<sup>-3</sup> and in London appears to have shown a small decline from around 4 to 3 µg m<sup>-3</sup>, which is fairly similar to that in Berlin. The data for Beijing show a similar magnitude of coarse particle increment. Non-exhaust particles are expected to comprise the main component of the coarse particle increment (Harrison et al., 2012) and as this is unaffected by emissions regulations, the lack of an obvious downward temporal trend is unsurprising.

In the case of NO<sub>2</sub> (Figure S5), the de-weathered roadside increment decreased from ~70 to ~50 µg m<sup>-3</sup> in London and Paris from 2009 to 2018. Beijing and Berlin show a much smaller increment in NO<sub>2</sub>. In the case of Beijing this is probably due to the lower emissions of a gasoline-fuelled light duty vehicle fleet (Yang et al., 2015) and in Berlin due to the clean air zone (Berlin.de, 2020). The Paris data appear to show an accelerated decline after 2013 which corresponded to the introduction of Euro VI standards (O'Driscoll et al., 2018). The temporal trend in the roadside increment in London is shown in Figure S6.

Urban increments (Figs. 2 and 3, Table 1) are often lower than the roadside increments. In particular, the NO<sub>2</sub> urban increment is more

than two times smaller than the roadside increment in London, Paris and Istanbul; NO<sub>x</sub> urban increment is 7 times lower than the roadside increment in London and more than two times lower in Berlin and Hong Kong (Table 1). In the case of PM<sub>2.5</sub>, the urban increment is substantially lower than the roadside increment in all cities except Istanbul. It is also found that PM<sub>2.5</sub> urban increment has declined from an average of around 5 µg m<sup>-3</sup> in 2009–2012 to values around 2.5 µg m<sup>-3</sup> in 2017–2018 in London (Figure S6).

Traffic exhaust particles consist primarily of organic (OC) and elemental (EC) carbon (Shi et al., 2000). Data from London enabled us to estimate the EC and OC contribution to roadside/urban increments. De-weathered black carbon (BC) at the heavily trafficked Marylebone Road site in central London is shown in Figure S7. BC decreased from 2009 to 2017 with an average decline of 0.57 µg m<sup>-3</sup> per year. In London, BC is generally taken as a surrogate for diesel exhaust emissions (this is possible because coal burning is banned and wood burning is a small seasonal source of particulate matter and has a low EC/OC ratio), and this shows the improvement effected by the implementation of diesel particle filters on new vehicles from 2011 onwards (ACEA, 2020). The EC and OC concentrations may be used to derive an approximate traffic PM<sub>2.5</sub> concentration, as explained in the Supplementary Material. This analysis in the Supplementary Material suggests that in London, the urban increment due to traffic is about one fifth of the roadside increment at Marylebone Road, and represents about 10% of the concentration of PM<sub>2.5</sub>.

Road traffic emissions also give rise to secondary pollutants, predominantly nitrate and secondary organic aerosol (SOA), the latter of which is formed from the volatile organic compounds (VOCs) from traffic exhaust. Predictions by numerical models are very uncertain due to the complexity of oxidation mechanisms, and the semi-volatility of both nitrate and some SOA components. For SOA, it is also difficult to attribute its mass to specific precursor VOC molecules (Hallquist et al., 2009; Ziemann and Atkinson, 2012). The reactions responsible for

formation of nitrate and SOA are typically rather slow and hence peak formation occurs downwind. Formation of nitrate from NO<sub>x</sub> emissions takes typically a few hours to one day, and therefore may have a limited impact upon local nitrate concentrations, except in large megacities. Table S1 indicates, based on national inventory data for NO<sub>2</sub> emissions, the respective contributions in London and Beijing. This suggests a modest contribution of road traffic to urban nitrate concentrations in London of 0.34 μg m<sup>-3</sup>, although if, for example, the London inventory were used to derive the percentage traffic contribution to NO<sub>x</sub>, the estimated traffic-related nitrate would be almost three times larger (0.83 μg m<sup>-3</sup>). The estimate from nitrate measurements (0.46 μg m<sup>-3</sup>) made by Abdalmogith and Harrison, 2006) lies between the two. Thus, the total (primary + secondary) contribution of road traffic to background PM<sub>2.5</sub> in London is about 13.4%. As expected, this is substantially lower than the roadside increment (48.6%, Table 1). There is a contribution of road traffic to the advected background pollution as measured by the rural data, but UK receptor modelling data indicate that this is small (Yin et al., 2015). Measurement and modelling based estimates also suggest a relatively small contribution of VOCs from road traffic to SOA, but the magnitude is uncertain (Gentner et al., 2017; McDonald et al., 2018; Lu et al., 2020; Jiang et al., 2019).

#### 4. Discussion

Although the twin site approach has been widely used to evaluate roadside and urban increments, it can be subject to artefacts if local sources other than road traffic affect one site but not the other, or if there are gradients in background pollution across an urban area. Also, comparing simultaneously measured data can be misleading in a period of rapidly changing concentrations, but this issue should not affect longer term averages from paired sites. Where suitable datasets existed (Beijing, Istanbul and Hong Kong), averages of several sites within a category were used for the calculation, as it is recognised that roadside increments are quite variable, dependent upon traffic volumes and local site characteristics such as canyon streets. There should be less variability in estimated urban increments for a city if the sites are carefully selected, and in London, for example, there was only minor variability in the urban background concentrations across the city, justifying the use of a single site. The roadside site selected for London is recognised as one of the most polluted roadsides in the city, while the site options available for Paris and Berlin were far fewer, and it is more difficult to judge representativeness. As a major aim of the study was to evaluate temporal trends in increments, site selection was heavily constrained by the need for long datasets at sites with a constant location.

The results reported above reveal that the road traffic contribution to PM concentrations, and especially PM<sub>2.5</sub>, has diminished very appreciably in the studied cities. In Beijing (Fig. 1) the increment between rural and urban concentrations is small and that between urban background and roadside is now also small in the case of PM<sub>2.5</sub>, coarse particles, SO<sub>2</sub> and CO. In London, the roadside and urban increments are a bigger percentage of the urban background concentrations, and represent 48.6% and 18%, respectively for PM<sub>2.5</sub>. In the case of NO<sub>2</sub>, the roadside increment is very large and the urban increment substantial. Taking account also of the contribution of road traffic to secondary nitrate and organic aerosol, road traffic continues to have an appreciable impact upon air quality, although the magnitude is highly variable in different cities.

The implications of this data analysis for air quality management depend critically upon the nature of the city in which management is required. It can be considered in three broad categories, although not all cities will exactly fit these descriptors.

##### 4.1. Developed world cities with strong controls on traffic emissions

London, Paris and Berlin fit this description and major progress has been made in the control of exhaust emissions of PM<sub>2.5</sub>. This source now

represents a relatively small component of overall PM<sub>2.5</sub> at urban background locations, and many roadside sites will exceed the WHO guideline (10 μg m<sup>-3</sup> annual mean) even when this source is eliminated. The largest gain from further control on road traffic emissions will be from a reduction in NO<sub>2</sub> concentrations, which remain in excess of the WHO guideline for NO<sub>2</sub> (40 μg m<sup>-3</sup> annual mean) at many near-traffic locations. Electrification of the vehicle fleet will have immediate benefits for NO<sub>2</sub> but will inevitably lead to higher concentrations of ozone especially at roadside (AQEG, 2009). It will also lead to a modest reduction in locally generated nitrate particles. Coarse particle emissions comprising mainly non-exhaust particles remain significant and there may be easy gains, for example, in relation to brake wear particles for which controls could be applied relatively inexpensively. There is also a need to develop better controls on road dust resuspension which is a significant source, particularly in the drier climates of the world. A move to battery electric vehicles will have only minor benefits for emissions of non-exhaust particles relative to modern internal combustion engine vehicles (Beddows and Harrison, 2020). As progressive further engineering controls on vehicle emissions give ever smaller benefits at ever higher costs, the ultimate answer for these cities must lie in a reduction in road traffic volumes.

##### 4.2. Less developed world cities with strong controls on traffic emissions, such as Beijing

Much has been achieved in Beijing to reduce emissions from the vehicle fleet including the adoption of ultra-low sulphur fuels and advanced exhaust aftertreatment technologies. The contribution of exhaust emissions from the largely petrol-fuelled light duty fleet is small in the case of PM<sub>2.5</sub>. The contribution of non-exhaust emissions to coarse particles is also small (Table 1). Thus, the gains from the removal of both the roadside traffic and urban increments would be small in the case of PM, the pollutant of greatest concern for public health, and reduction of other local and regional sources of particulate matter should be a priority (Wu et al., 2020). There is still an important contribution of traffic to NO<sub>2</sub> concentrations and thus to regional nitrate formation. Similar to those developed cities such as London, electrification of the vehicle fleet will lead to a large reduction in NO<sub>2</sub> concentrations and modest reduction in nitrate aerosols but this may cause an increase in ozone without further control on VOC emissions (AQEG, 2009). Consequently, policy development needs high quality chemistry-transport modelling to chart a course to cleaner air quality with respect to all pollutants, so as to avoid unintended consequences. Hong Kong and Istanbul appear also to fit this class of city (Table 1).

##### 4.3. Less developed world cities with poor controls on traffic emissions

Data in these cities are not readily available but their vehicle fleet is both old and poorly maintained and there has been as yet no transition to ultra-low sulphur fuels. Traffic pollution can be a major source of a number of pollutants, Delhi being an example (Pant et al., 2015; Kumar et al., 2015). Based on historical data in western countries (Figures S3, S5 and S6), there can be major air quality improvements resulting from an upgrade to the motor-vehicle fleet and an introduction of higher quality ultra-low sulphur fuels.

#### 5. Data availability

Data supporting this publication are openly available from the UBIRA eData repository at <https://doi.org/10.25500/edata.bham.00000420>.

#### CRedit authorship contribution statement

**Roy M. Harrison:** Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Writing - original draft,

Writing - review & editing. **Tuan Van Vu:** Data curation, Formal analysis, Investigation, Software, Visualization. **Hanan Jafar:** Formal analysis, Investigation, Visualization. **Zongbo Shi:** Funding acquisition, Project administration, Supervision, Writing - review & editing.

## Declaration of Competing Interest

The authors declared that there is no conflict of interest.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2020.106329>.

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