Supplementary Material

S.1 Scenarios for particle number emission estimates

Two scenarios, representing the worst and the best, are considered for the analysis.

Business as usual (BAU): This is a base case scenario in which no policy interventions are considered. After the construction of this scenario (Section S.2), the BAU results in 5.40 and 5.58 times increases in total vehicle population and total VKT, respectively, in 2030 from the 2010 levels. The LDVs, buses and 3Ws registered in Delhi after 2006 are assumed to running on CNG, except the vehicles coming in from the outside states and passing through Delhi. The phasing out of vehicles after the retirement age of 15 years (public) and 17 years (commercial), together with complete removal of 2-stroke 2Ws by 2015, is considered as per Delhi Government norms. The base line data for different traffic between 1991 and 2006 is projected for future years until 2030. Note that the vehicle data between 1991 and 2006 represent actual registrations of vehicles in Delhi. Future growth in vehicles is estimated based on the socio-economic analysis which provided strong correlations between the GDP for Delhi and total number of different vehicle type (see Section S.2 for details).

Best estimate scenario (BES): The BES considers every possible reduction in nanoparticle emissions due to interventions by transport and emission control policies and infrastructural development for road transport. After the final construction, the BES ends up in 3.09 and 4.03 times increase in total vehicle population and VKT, respectively, in 2030 from the 2010 levels (Section S.2). Other considerations include hypothetical implementation of emission control technologies, changes in fuel and vehicle types, improved vehicle speeds due to implementation of multi mode mass transit system (MRTS) and BRT corridors, phasing out of both public and commercial vehicles after a short retirement age and complete phasing out of 2-stroke 2Ws by 2012, as suggested by CAI [1] (see Section S.2 for details).

S.2 Methodology

Since there is no standard data base available for vehicle population in Delhi, we have made estimations using the data published in relevant studies and making realistic assumptions under different scenarios, as explained below and summarised in Fig. S.1:
Total vehicle population in BAU \((N_{v,BAU}) = \) Number of registered road vehicles + External vehicles coming in and passing through the city – Phased out old vehicles\(^1\)

Total vehicle population in BES \((N_{v,BES}) = N_{v,BAU} – \) Vehicles off the road due to MRTS and Bus Rapid Transit System (BRT) in Delhi

### S.2.1 Base vehicle population

Following methodology is adopted for modelling the vehicle population in different categories for future years. Firstly, data for registered vehicles in Delhi between 1991 and 2006 are complied [2]. Future growth in vehicles is estimated based on the socio-economic analysis between the annual gross domestic product (GDP) growth and total cumulative number of annually registered vehicles for the years between 2001 and 2006. This trend was then extended to project vehicle population after 2006 by assuming a 10% annual growth in GDP that is suggested by Planning Commission of Delhi. The estimated average annual growth was found to be 10.8, 13.3, 13.6, 6.7, 8.2 and 9.5% for 3Ws, taxis, buses, goods vehicle (i.e. LDVs and HDVs), cars and jeeps, and 2Ws, respectively. Our estimates are on a bit higher side than those suggested by Murty et al. [3] for 3Ws (8%), taxis (5%), buses (7%), cars and jeeps (10%), and 2Ws (9.8%) due to consideration of a little higher GDP growth than anticipated in past years.

#### S.2.1.1 \(N_{v,BAU}\)

After projecting the vehicle population until 2030, it was then corrected to consider the population of external vehicles coming into (and passing through) the city [4]. This was applied considering the past ratios of internal to external vehicle populations, as specified by Mashelkar et al. [4] for the year 2001–2005. These were 27.06, 8.41, 24.35, 27.06, 47.83, 57.76 and 57.76% for cars and jeeps, 2Ws, 3Ws, taxis, buses, LDVs and HDVs, respectively.

The total vehicle population was then further divided into fuel specific vehicle categories. For cars and Jeeps, the proportion was assumed as 85 and 15% for diesel and gasoline driven vehicles, respectively [5]. Data for CNG driven vehicles for cars and jeeps (and other vehicles) between 1997 and 2003 was collected from different sources [5-10] and their growth rate for future years was estimated accordingly. The quantity of CNG driven vehicles was then taken out equally from both the gasoline and diesel driven cars and jeeps vehicles.

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\(^1\)For BAU, retirement age for vehicles is considered as per the Delhi Government norms [21, 23, 34]. For the BES, phasing out age of different vehicles are considered as per the Clean Air Initiative for Asian Cities [1, 13].
For 2Ws and HDVs, it was assumed that all the vehicles are running on the gasoline and diesel, respectively. For 3Ws, it was assumed that they either run on gasoline or on CNG. Data for CNG driven 3Ws was collected from the same sources [5-10] to those for cars and jeeps and estimated their growth rate for future years. Gasoline driven 3Ws were then adjusted by subtracting the CNG driven 3Ws from them. Share of taxis was assumed as 25 and 75% for diesel and gasoline [5]. The same technique, as used to estimate the CNG driven cars and jeeps, was applied to estimate the CNG, gasoline and diesel driven taxis. Buses were assumed to be driven by diesel and CNG; the similar method adopted for 3Ws provided their share in diesel and CNG category. LDVs were assumed as driven by CNG and diesel. It was also assumed that all new registered LDVs must be CNG after 2006 [9] and the numbers of diesel driven LDVs were adjusted accordingly after 2006.

The next step was to take out the phasing out of vehicles from the vehicle fleet. Following the Delhi Government norms introduced in 2001–2002 [4, 9, 11], the number of old–aged retired vehicles has been subtracted from the entire data set by assuming the retirement age as 17 years for private vehicles and 15 years for commercial vehicles. Furthermore, all the 2–stroke 2–wheelers are assumed to be phased out by 2015, following a ban by the Delhi Government in 2000 [12].

S.2.1.2 \( N_{v,BES} \)

For estimating the \( N_{v,BES} \), the \( N_{v,BAU} \) values were first adjusted by modifying the retirement age as 15, 10, 10 and 8 years for buses, cars, 3Ws and 2Ws, respectively, as suggested by the Clean Air Initiative for Asian cities [1, 13]. All 2–stroke motorcycles (i.e. 2Ws) are assumed to be phased out by 2012 therefore 2–stroke 2Ws are removed from the vehicle count after 2012 [13]. The vehicle population is then further adjusted due to the implementation of MRTS in Delhi. This reduction in vehicle population between 2006 and 2030 is considered for private vehicles such as cars and jeeps, motorcycles and buses, as suggested by Murty et al. [3]. It is also assumed that other vehicles like taxis and 3Ws are on the road by choice and will not be diverted by the MRTS. It is also considered that the MRTS will increase the speed of road vehicles by about 25% after the completion of Phase II in 2011 due to decongestion [3]. Furthermore, a total of 26 BRT corridors are planned that will cover a total length of 310 km by the year 2020 in three phases (I: 2005–2010; II: 2010–2015; III: 2015–2020). This is expected to decrease the travel time by 15–30% [14] by increasing the speed of buses by 15–30% [15]. We have not considered any reduction in private vehicles due to the BRT corridors, assuming that the number of buses will remain the same but will run at a much higher speed. Consequently, we have
assumed a 25% increase in speeds of all vehicles after 2011 due to MRTS in our calculations, and a further increase of 20% in the speed of buses after 2020 due to BRT.

Furthermore, a shift of vehicle fuels is considered in the light of the following assumptions: (i) it is considered that all the taxis are running on CNG after 2006, except the external ones which are assumed to have 75 and 25% share for gasoline and diesel, respectively [5], (ii) all the buses and LDVs (either internal or external) are running on CNG after 2006. The last assumption made under the BES scenario is that the all the diesel vehicles (both internal and external) are retrofitted with a DPF by 2020 which will presumably be necessary for the majority of vehicles not complying with the Euro IV emission standard in London low emission zone from January 2012.

Fig. S.1. Vehicle population and VKT travelled in Delhi under both BAU and BES scenarios.

S.2.2 Vehicle kilometre travelled (VKT)

For both scenarios, the annual VKT for each vehicle category are estimated by multiplying the VKT/day with the total number of days in a year (Fig. S.1).

- **BAU**: Total VKT per day \( VKT_{BAU} = N_{v,BAU} \times \text{VKT by each vehicle type per day} \)
- **BES**: Total VKT per day \( VKT_{BES} = N_{v,BES} \times \text{VKT by each vehicle type per day} \)

The VKT/day were assumed as 41 (cars), 27 (2Ws), 110 (3Ws), 164 (Buses), 82 (HDVs and taxis) and 110 (LDVs) [3, 16]. Once the total VKT were calculated, these were then further divided into three periods (peak, off–peak and free flow; see details in Section S.2.3) and the PNEF were chosen
according to the corresponding vehicle speeds during these periods. It was estimated from our previous work that the total VKT travelled during morning and evening peaks, off–peak and free flow periods were 53, 40 and 7% respectively [17].

For the BAU, average vehicle speeds during peak hours were assumed to be 26 (cars and jeeps, taxis), 27 (2Ws), 23 (3Ws), 17 (buses), 25 (HDVs) and 10 km h$^{-1}$ (LDVs) [18]. An increase of 11% from peak hours is considered for off peak hours [18-19]. During the free flow traffic conditions, which usually occurs in night, the maximum speed was limited to 60 km h$^{-1}$ which was assumed to be achieved by all vehicles, except the 3Ws assumed to be running at an speed of 48 km h$^{-1}$ [20]. Under the BES, average vehicle speed is taken as the vehicle speeds during the BAU plus the increase due to infrastructural development as suggested in Section S.2.1.2.

S.2.3 Particle number emission factors (PNEF)

Table 4 our recent review article (Kumar et al. [21]) on the ‘dynamics and dispersion modelling of nanoparticles from road traffic in the urban atmospheric environment’ presents a comprehensive review of the PNEF studies carried out in the last 2 decades for individual types (heavy duty vehicles, HDVs, light duty vehicles, LDVs, cars (petrol–fuelled, diesel fuelled), buses (diesel–fuelled, compressed natural gas fuelled, petrol and light petroleum gas fuelled spark ignited vehicles, and two–wheelers), and mixed fleet of road vehicles in varying operational conditions. Table S.1 summarises the PNEFs presented in the Table 4 of Kumar et al. [21] for deriving the nanoparticle emission estimates in megacity Delhi in both scenarios, according to the speeds of the individual vehicle type. For example, under the BAU, these are selected for different vehicle types depending on the vehicle speeds during the three designated time periods: (i) morning and evening peaks (0800–1200h; 1600–2000h), morning and evening off–peaks (0600-0800; 1200–1600h; 2000–2200h) and free flow (2200–06:00).

Under the BES, different values of PNEFs have selected from Table S.1 due to the change in fuel types, speeds and diesel particulate trap (DPF) since it is assumed that all diesel vehicles (either internal or external) are retrofitted by the diesel particulate trap (DPF) by 2020 which is going to be legally effective, for example in London, from January 2012. Retrofitting of DPFs to diesel vehicles also meant that the particle number emissions can be reduced up to two orders of magnitude for the same diesel vehicles compared with the non–DPF treatment [22]. As discussed in S.2.1.2, the implementation of the MRTS and BRT will enhance the speeds of certain type of vehicles, meaning that the selected PNEFs will be positively affected by increasing vehicle speeds.
Table S.1. Summary of PNEFs; these are derived from the table presented in Kumar et al. [21]; these are used for estimating ToN emissions in our study. Vehicle speeds are broadly categorised in two ranges to accommodate the variability in speeds during peak and off–peak hours. Numbers in parenthesis shows the minimum and maximum values of PNEF with average values in front of them.

<table>
<thead>
<tr>
<th>Vehicle type</th>
<th>Speed (km h⁻¹)</th>
<th>PNEF (# Veh⁻¹ Km⁻¹)</th>
<th>PNEF (# Veh⁻¹ Km⁻¹) with DPF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cars (diesel)</td>
<td>&lt;50</td>
<td>1.12 [0.44–1.8] × 10¹⁴</td>
<td>d6.71 × 10¹⁰</td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>5.27 [0.57–9.97] × 10¹⁴</td>
<td></td>
</tr>
<tr>
<td>Cars (gasoline)</td>
<td>&lt;50</td>
<td>2.3 [0.12–4.5] × 10¹³</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>2.8 [0.18–5.3] × 10¹³</td>
<td></td>
</tr>
<tr>
<td>Cars (CNG)a</td>
<td>&lt;50</td>
<td>2.25 [1.5–3] × 10¹⁰</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>5.0 [0.006–10] × 10¹²</td>
<td></td>
</tr>
<tr>
<td>HDVs (Diesel)</td>
<td>&lt;50</td>
<td>2.95 [0.39–5.5] × 10¹⁵</td>
<td>e2.95 [0.39–5.5] × 10¹³</td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>4.04 [0.78–7.3] × 10¹⁵</td>
<td>e4.04 [0.78–7.3] × 10¹³</td>
</tr>
<tr>
<td>LDVs (Diesel)</td>
<td>&lt;50</td>
<td>1.27 [0.8–1.74] × 10¹⁴</td>
<td>d6.71 × 10¹⁰</td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>4.05 [1.2–6.9] × 10¹⁴</td>
<td></td>
</tr>
<tr>
<td>LDVs (CNG)a</td>
<td>&lt;50</td>
<td>2.25 [1.5–3] × 10¹⁰</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>1.25 [0.006–25] × 10¹²</td>
<td></td>
</tr>
<tr>
<td>Buses (Diesel)</td>
<td>&lt;50</td>
<td>3.87 [1.38–6.36] × 10¹⁴</td>
<td>d1.16 [0.59–1.73] × 10¹³</td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>9.6 [1.2–18] × 10¹⁴</td>
<td></td>
</tr>
<tr>
<td>Buses (CNG)</td>
<td>&lt;50</td>
<td>2.25 [1.5–3] × 10¹⁰</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>1.25 [0.0018–2.5] × 10¹³</td>
<td></td>
</tr>
<tr>
<td>2Ws b</td>
<td>&lt;50</td>
<td>0.15 [0.1–0.3] × 10¹³</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>5.27 [0.57–9.97] × 10¹⁴</td>
<td></td>
</tr>
<tr>
<td>3Ws (gasoline)c</td>
<td>&lt;50</td>
<td>2.3 [0.12–4.5] × 10¹³</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>2.8 [0.18–5.3] × 10¹³</td>
<td></td>
</tr>
<tr>
<td>3Ws (CNG)c</td>
<td>&lt;50</td>
<td>2.25 [1.5–3] × 10¹⁰</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&gt;50</td>
<td>5.0 [0.006–10] × 10¹²</td>
<td></td>
</tr>
</tbody>
</table>

a Due to lack of data, emissions for CNG driven cars and LDVs are assumed same as those from CNG driven buses at low and medium engine loads, respectively.

b The chosen values are based on a limited data set, but these compare well with the findings of Etisa et al. [23]. They reported that number concentration and size distribution of the particles emitted by 2–stroke 2Ws are roughly in the range of 4–stroke diesel engines; however, the nature of the particles would be different.

c No suitable data could be found in the literature, therefore it is assumed that gasoline and CNG driven 3Ws emit similar to gasoline and CNG driven cars, respectively.

d Source EIG [24]

e Assumed that the DPFs will reduce 99% emissions compared with non–DPF diesel engines [22].
S.3 Formulation of a simplified box model

The simplified model derived for a street canyon as a part of our early work [25] is modified for deriving the ToN concentrations \(C\) in megacity Delhi. Formulation of the box model assumes that the whole city acts as a control volume (box), and that the air in the box is well mixed and uniform in concentration and there is a uniform wind velocity \(U_r\) in cm s\(^{-1}\) flowing along the \(x\)-direction (i.e., horizontally), and an exchange wind velocity \(\bar{w}\) in cm s\(^{-1}\) in the \(z\)-direction (i.e., vertically).

We assume that the box has width \((W)\) and extends up to the city mixing height \((H_m)\) and that there is background pollution \((C_b\) in \(#\text{ cm}^{-3}\) being advected (horizontally and vertically) in and out of the box and that the vehicles in the box are the only uniformly distributed line source that generates \(Q\) (particle number flux) \(#\text{ cm}^{-2}\text{ s}^{-1}\); total particle numbers in the box are assumed to be conserved. Also, the removal due to deposition and gravitational settling are assumed to be negligible, and that there is no change in PNCs through transformation processes in the box. However, the effect of transformation processes (e.g. dry deposition, coagulation) is taken into account separately, as described in Section S.4.

Then the conservation of mass for this pollutant gives for its concentration \(C\) (\(#\text{ cm}^{-3}\)) in the box as;

\[
LWH_m \frac{\partial C}{\partial t} = LWQ + [C_b - C]U_r H_m W + [C_b - C] \bar{w} WL \quad (S-1)
\]

\[
\text{Change in } C \text{ with time} = \frac{\partial C}{\partial t} = \text{Source term} + \text{In – Out (Through horizontal advection)} + \text{In – Out (Through vertical exchange)}
\]

\[
\frac{\partial C}{\partial t} = \frac{Q}{H_m} + \frac{[C_b - C]U_r}{L} + \frac{[C_b - C] \bar{w}}{H_m}
\]

or

\[
\frac{\partial C}{\partial t} = \frac{Q}{H_m} + C_b \left[ \frac{U_r}{L} + \frac{\bar{w}}{H_m} \right] - C \left[ \frac{U_r}{L} + \frac{\bar{w}}{H_m} \right] \quad (S-2)
\]

For Steady state conditions:

\[
C = \frac{Q}{H \left[ \frac{U_r}{L} + \frac{\bar{w}}{H_m} \right]} + C_b \quad (S-3)
\]

The first denominator term \((U_r/L)\) term in Eq. (S–3) represent horizontal transport while the second denominator term \((\bar{w}/H_m)\) indicates vertical transport [26]. The \(C_b\) is ignored due to the unavailability of the data, giving us only the vehicle derived concentrations. The vertical transport term is also ignored considering that the concentrations are well mixed in the assumed box which has a mixing height \((H_m)\) of about 200 m; details for its estimation are provided below. The vertical profile of the horizontal wind velocity is assumed as logarithmic, but considered separately through Eq. (S–4) for taking into account
the local vertical exchange between the above–and urban–canopy flows. European Union directive 1999/30/EC suggests sampling heights between 1.5 and 4m above road level, and up to 8m above road level under specific local circumstances [27]. Eq. (S–4) allows estimating the $U_r$ ($0.44\pm0.25$ m s$^{-1}$) at about 2.0 m ($z$) which is the representative of typical height of urban monitoring stations at roadsides.

\[
U_r = \frac{u^*}{\kappa} \ln \left( \frac{z - d}{z_0} \right) \tag{S–4}
\]

where $u^*$ is surface friction velocity which is estimated as 0.26 m s$^{-1}$ based on the known annual mean $U_r$ ($2.14$ m s$^{-1}$) at about 15 m height; $\kappa$ (=0.4) is a constant; $d$ (=1m) is the zero displacement height; $z_0$ (=0.5 m) is surface roughness length [28]. Considerations of the above assumptions transform the Eq. (S–3) into the following:

\[
C = \frac{Q}{H_m \left[ \frac{U_r}{L} \right]} \tag{S–5}
\]

where $Q$ is the net particle number flux out of the canyon (i.e., the net number of particles passing through unit upper surface area, $A_x$ in cm$^2$, in unit time) can be written as [29]:

\[
Q = \sum_{x=1}^{n} E_{x,i-j} T_x \frac{T_x}{A_s} = \frac{ToN}{A_s} \tag{S–6}
\]

where $E_{x,i-j}$ is the particle number emission factor in # veh$^{-1}$ cm$^{-1}$ in any size range of a vehicle in $x$ class, $T_x$ is the number of vehicles per second of class $x$, and ToN is in # s$^{-1}$. Substituting the value of $Q$ into Eq. (S–5) produces:

\[
C = \frac{ToN}{A_s H_m \left[ \frac{U_r}{L} \right]} = \frac{ToN \times L}{A_s \times H_m \times U_r} \tag{S–7}
\]

where $L$ (=47.53 km) is the assumed length of the Delhi city which is derived from the Fig. S.2. For deriving the length and breadth for our box model, the rectangular shape is considered which has total equivalent surface area of Delhi as 1483 km$^2$. The predominant wind direction is taken as north–west as found by Mehra at al. [30] and Saksena et al. [31] for Delhi. The model assumes uniform ToN concentrations in the box but to take into account the vertical profile of the horizontal wind velocity that is needed to estimate ToN concentrations at the roadside (~2.0 m), Eq. (S–7) of box model is further modified by replacing the $U_r$ with Eq. (S–4). This model disregards any transformation processes but the resultant ToN concentrations are corrected separately for transformation losses (see Section S.4).

Since we needed the mean wind speed for Eq. (S–7) and could not locate a reliable published data covering our study period, we analysed the online hourly averaged data available at
These data are continuously taken on an hourly basis at Safdarjung (New Delhi) since 1996; monitoring station is located approximately in the centre of Delhi at about 15 m above the ground level. As seen in Fig. S.3, average $U_r$ between the available periods (i.e. 1996 and 2010) was found to be reasonable constant (i.e. $2.14 \pm 1.21 \text{ m s}^{-1}$). We used the wind speeds shown in Fig. S.3 for the available years, and an average value ($2.14 \pm 1.21 \text{ m s}^{-1}$) for the remaining years.

Fig. S.2. Aerial map of megacity Delhi [12], showing an assumed box and prevailing wind direction from the north-west (figure not to scale). The rectangular box indicates assumed length and width for our box model giving a total equivalent area of 1483 km$^2$.

The $H_m$ is the height of well–mixed layer which is typically encountered under significant convective conditions (i.e. heat, mass and momentum transfer between the local earth surface and the atmosphere) [32]. The $H_m$ embeds major part (50–80%) of the convective atmospheric boundary layer [33] depending on the atmospheric turbulence (buoyancy and wind shear). In Delhi, $H_m$ can vary between few 10’s and 100’s of meters during the stable (night) and unstable (day) conditions, respectively. In fact, the values of $H_m$ are location specific since they can vary depending on the convective activity. For example, Ketzel and Berkowicz [34] assumed the typical values of $H_m$ as 500 m while Gighagen et al. [35] assumes it as 100 m in their works. On the other hand, Britter and Hanna [28]
suggest that it could be up to the twice the mean height of buildings. Considering these variability’s and rather than choosing a random value, we made inverse calculations using the Eq. (S–7) for estimating the $H_m$. For this purpose, we choose the results of an independent study (i.e. Monkkonen et al. [36]) that measured ToN concentrations in Delhi. The following information was feed into the Eq. (S–7): (i) average ToN concentrations measured between 26 October 2002 and 9 November 2002 by Monkkonen et al. [36] as $6.28 \times 10^4$ cm$^{-3}$, (ii) $U_r$ as 1.07 m s$^{-1}$, which is computed for the duration between 26 October 2002 and 9 November 2002 using the online wind speed data available at: http://www.wunderground.com/history/, (iii) $L$ and $A_s$ are considered as described above for Eq. (S–7), and (iv) ToN emissions for the year 2002 are estimated as $4.22 \times 10^{17}$ # s$^{-1}$ using the methodology described in Section 2.3 of the article. Substituting these values in Eq. (S–7) gives $H_m$ as 200.66 m.

![Annual mean wind speed](image)

**Fig. S.3.** Hourly averaged annual mean wind speed for Delhi.

### S.3.1 Summary of values used for estimating ambient and roadside ToN concentrations

The section below describe the variables and their values used in Eq. (S–7) for converting the annual ToN emissions into the hourly averaged *roadside* and *ambient* ToN concentrations [29]. ToN is in # s$^{-1}$ and $L$ (=47.53 km) is the assumed length of the Delhi which is derived from the Fig. S.2. $H_m$ is the mixing height which is computed as 200 m (see Section S.3); $Q$ is particle number flux (# cm$^{-2}$ s$^{-1}$)
which is defined as the net number of particles passing through per unit surface area ($A_s$; in cm$^2$) per unit time; $U_r$ (cm s$^{-1}$) is the hourly average synoptic (i.e. above urban canopy) wind speed as described in Section 3.2. The $A_s$ depends on the particle production rate within that area by the sources (e.g. road vehicles in our case) and any influence of conversion processes which are ignored here. Two different values of the $A_s$ are considered under the following assumptions: (i) $A_s$ is equivalent to the entire city area (1483 km$^2$) assuming the whole city as a uniformly distributed area source for mimicking ambient concentrations, and (ii) $A_s$ is equivalent to surface area covered by roads (including roadside land) for mimicking on–road or roadside concentrations; $A_s$ is taken as 21% of total Delhi area [37] up to 2005 plus a further equivalent increase in area for an additional 310 km road length between 2005–2010, 2010–2015 and 2015–2020 due to BRT corridors. As described in Section S.3, average $U_r$ is taken as 2.14 (0.92–3.35, representing lower–upper standard deviation) m s$^{-1}$ for estimating corresponding ambient ToN concentrations (Table 3); these values are derived from Fig. S.3 (see Section S.3 for details). The $U_r$ within the urban canopy is typically smaller than the above–canopy flows [28], meaning that roadside concentrations will be larger than the ambient concentrations. For estimating roadside concentrations at about 2.0 m height, which represent typical height of monitoring stations in urban areas, the $U_r$ explained through Eq. (S–4) is modified by substituting this by the $U_{2.0} = \frac{u^*}{\kappa} \ln \left( \frac{2 - d}{z_0} \right)$

As described in Section S.3, $u^*$ (=0.26 m s$^{-1}$) is surface friction velocity; $\kappa$ (=0.4) is a constant; $d$ (=1 m) is the zero displacement height; $z_0$ (=0.5 m) is surface roughness length [28].

S.4 Effect of transformation processes on ToN concentrations

Transformation processes (e.g. coagulation, nucleation, condensation, evaporation, and deposition) show varying effects on particles of different sizes [21]. Information on particle size distributions for Delhi is not available through our estimates. We have therefore assumed the measured size distributions for Delhi by Monkkonen et al. [36] for making approximations of ToN losses due to various transformation process. As discussed in Section 3.4, we only considered the effects of coagulation, deposition and nucleation for correcting our estimated ToN concentrations; methodology to estimate them is shown in the text below.

The following equation is used to estimate the losses of ToN due to coagulation [38]:

$$ToN_{\text{AfterCoag}} = \frac{ToN_{\text{Initial}}}{1 + \frac{ToN_{\text{Initial}}}{K_t}} \quad (S–9)$$
where \( t \) is time and \( \bar{K} \) is the average polydisperse coagulation coefficient between \( k \) size intervals (i.e. nucleation, Aitken and accumulation), as defined in Eq. (S–10).

\[
\bar{K} = \sum_{i=1}^{k} \sum_{j=1}^{k} K_{i,j} f_i f_j \quad \text{and} \quad K_{i,j} = \frac{\pi(d_i D_i + d_j D_j + d_i d_j)}{2} \beta
\]

where \( f_i \) and \( f_j \) are fractions of the ToN concentrations in the \( i^{th} \) and \( j^{th} \) intervals and \( D \) (cm\(^2\) s\(^{-1}\)) is their diffusion coefficient. Since the values of ‘\( K_{i,j} \)’ are particle size dependent, we have assumed a trimodal distribution (nucleation, Aitken and accumulation) as given by Monkkonen et al. [36] for Delhi. They found geometrical mean diameters (GMD) in nucleation, Aitken and accumulation modes as about 11, 44 and 147 nm, respectively, with ToN concentration distributions about 8, 58 and 34\%, respectively (Table S.2). For the simplicity, we assumed two broad modes (i.e. one, the combination of both nucleation and Aitken modes, and the other accumulation mode) for approximating the value of \( K \). Using concentrations distributions in each mode and assuming that all the particles in each mode has size equal to their GMDs but polydisperse coagulation occurs among these modes, the value of \( K \) is estimated as \( 3.11 \times 10^{-10} \) cm\(^3\) s\(^{-1}\). The value of \( \beta \) was computed as 0.65; this is the particle diameter dependent weighted average transitional correction factor (see Table 12.1 of Hinds [38]). Resulting particle losses due to coagulation, derived under the different scenarios, are presented in Table S.3.

**Table S.2.** Estimates of GMD and ToN concentrations and proportions are based on the sample data taken from Monkkonen et al. [36] and Laakso et al. [39]; deposition velocity \( (v_{d,i}) \) are derived from Fig. S.3. Following data are used for approximating particle losses due to coagulation and deposition.

<table>
<thead>
<tr>
<th>Modes</th>
<th>GMD (nm)</th>
<th>( v_{d,i} ) (cm s(^{-1}))</th>
<th>ToN (# cm(^{-3}))</th>
<th>( f ) (ToN in %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nucleation</td>
<td>11</td>
<td>0.47</td>
<td>( 5.15 \times 10^3 )</td>
<td>0.08</td>
</tr>
<tr>
<td>Aitken</td>
<td>44</td>
<td>0.09</td>
<td>( 37.97 \times 10^3 )</td>
<td>0.58</td>
</tr>
<tr>
<td>Accumulation</td>
<td>147</td>
<td>0.02</td>
<td>( 22.68 \times 10^3 )</td>
<td>0.34</td>
</tr>
</tbody>
</table>

Furthermore, losses in ToN concentrations due to dry deposition of particles are approximated using the following equation [34, 40]:

\[
ToN_{AfterDep} = ToN_{Initial} - ToN_{Initial} \left( \frac{v_{d} T_d}{H_m} \right)
\]

Substituting the dry deposition velocity, fractions \( (f) \) of ToN and ToN concentrations in nucleation, Aitken and accumulation modes in Eq. (S–11) gives:
where $T_d$ is the deposition time which is defined as $L/U_r$. Size dependent dry deposition speed ($v_d$) was computed using the deposition model of Seinfeld and Pandis [41] after feeding the typical values of input parameters for urban conditions. For instance, aerodynamic resistance ($r_a$) was taken as 33 s cm$^{-1}$ [34, 42] and $u^*$ was estimated using the Eq. (S–4) as 0.26 cm s$^{-1}$ for our conditions. The resulting size dependent $v_d$ is plotted against the particle diameter in Fig. S.4. As expected, the values of $v_d$ decreases and then increases with increasing size due to larger diffusion coefficients for smaller particles and the settling velocity dominating the larger size range (Fig. S.4). Table S.2 shows the chosen values of $v_d$ from the Fig. S.4, for the same GMDs as taken for coagulation estimates, in order to estimate the ToN losses due to dry deposition.

\[ ToN_{AfterDep} = \frac{ToN_{Initial} T_d}{H_m} \left( v_{d(Nuc)} ToN_{Nuc} f_{Nuc} + v_{d(Air)} ToN_{Air} f_{Air} + v_{d(Acc)} ToN_{Acc} f_{Acc} \right) \]  

(S–12)

According to a Delhi study [36], the formation rate of 3 nm particles of the observed events varied from 3.3 to 13.9 cm$^{-3}$ s$^{-1}$ with the growth rate varying from 11.6 to 18.1 nm h$^{-1}$; these growth and formation rates represent typical urban conditions [43]. The rate of formation observed by Shi et al. [44]
for 3 nm particles in Birmingham (UK) ranged between 5 to 50 nm with a growth rate of about 4 nm h$^{-1}$; these encompasses the rate of formation observed for Delhi. Since rates of new particle formation are available for Delhi [36], we used 3.3 cm$^{-3}$ s$^{-1}$ for making an conservative estimate for the production of nucleation mode particles. However, nucleation events were found rarely in London and Birmingham. For instance, these were observed only about 12 days out of total 232 days of monitoring (i.e. 20% of time) in Birmingham [45]. Given the very high background particle loading in Delhi, and a larger condensation sink, it is unlikely to have countable effect of nucleation process. For justifying this assumption, it is assumed that there is an half an hour nucleation event in 24 hour for 20% of days in a year. The ToN produced during nucleation event is then equally distributed over 24 hour period by adding this fraction to the hourly averaged concentrations. Percent changes in ToN concentrations due to nucleation in different scenarios are shown in Table S.3 which are, as expected, modest.

Temperature affects the formation of atmospheric nanoparticles [46-47], and hence the estimates of PNEFs, appreciably. We have however discarded its effect assuming that the PNEFs used for our calculations are estimated by numerous studies (see details in Kumar et al. [21]) during a range of temperature conditions which encompasses through a hugely variable ambient temperature of Delhi over a year. For instance, the extreme temperatures could be as low as –0.6ºC during December–January and as high as 47.2 ºC during May–June.

Table S.3. Effect of transformation processes on ambient ToN concentrations.

<table>
<thead>
<tr>
<th>Year</th>
<th>Coagulation</th>
<th>Dry Deposition</th>
<th>Nucleation</th>
<th>Net effect</th>
</tr>
</thead>
<tbody>
<tr>
<td>2010</td>
<td>–3.53</td>
<td>–10.64</td>
<td>0.18</td>
<td>–13.99</td>
</tr>
<tr>
<td>2030–BES</td>
<td>–0.21</td>
<td>–10.64</td>
<td>0.04</td>
<td>–10.81</td>
</tr>
</tbody>
</table>

S.5 Estimates of daily mortality from exposure to PNC in Delhi

For calculating the numbers of deaths brought forward (total mortality) as a result of exposure to airborne nanoparticles as described by particle number count, it is necessary to use an exposure–response coefficient which relates a change in particle number count to the number of associated deaths. Whilst these are abundant in the literature for the effects of exposure to PM$_{10}$ concentration, they are almost non–existent for particle number. The very few values available include that reported by Atkinson et al. [48] from a time series study conducted in London, and by Stolzel et al. [49] for Erfurt,
Germany. They report coefficients relating particle number count to total mortality, cause–specific mortality from respiratory and cardiovascular diseases and in the former paper hospital admissions for respiratory and cardiovascular diseases. As reliable data were not available from Delhi for either cause–specific mortality in the general population or hospital admissions, the calculation has been conducted only for the effects on total mortality. It is very likely that the criteria for hospital admission differ substantially between Delhi and London and consequently application of European coefficients for hospital admissions to Delhi would be inappropriate. Other researchers (e.g. Gurjar et al. [50]) have used hospital admission rates from developed countries as a surrogate for the lacking data but this was not felt to be appropriate for this study. Our calculations assume that death rates for 2008 are applicable to Delhi’s population in 2010 and 2030 and that the exposure–response coefficient remains unchanged.

The population data for the calculation were derived from the World Health Organisation [51] and the mortality rate from the Annual Report on Registrations of Births and Deaths for Delhi [52]. The exposure–response coefficient from Atkinson et al. [48] is a value from lag 1 (i.e. the day after exposure occurred) from a time series study. The coefficients for Stolzel et al. [49] are for a lag 4 and from a polynomial distributed lag model also derived from a time–series study. These express only the effects of recent exposure upon daily mortality rates. The public health impacts of exposure to particulate matter as expressed by PM$_{2.5}$ are much greater for the chronic impacts in which life–long exposure leads to premature mortality than to the acute impacts of daily exposures. However, no comparable data exists for particle number exposures and consequently no calculation of chronic effects can be made at this time. Table S.4 shows the summary of the data which is used to estimate the total mortality by putting them in the following expression:

$$\text{Death brought forward (DBF)} = \text{Population} \times \text{Mortality rate} \times \text{PNC} \times C_{E-R} \quad (S–13)$$

In Eq. (S–13), PNC is the particle number concentrations and the $C_{E-R}$ is exposure–response coefficient for nanoparticle exposure that is estimated using the relative risk (RR) values given in Table S.4 (i.e. $C_{E-R} = RR–1$). Atkinson et al. [48] found a significant relationship between total mortality and particle number count at lag 1 for which the relative risk and 95% CI is 1.013 (1.005–1.022) per 10$^4$ PNC. Alternatively, two significant coefficients relating total mortality to the PNC between 10 and 100 nm were estimated by Stölzel et al. [49]. Taking account of the inter–quartile range, the coefficients are for lag 4 with 95% CI are 1.030 (1.003–1.056) and 1.043 (1.014–1.072) for the polynomial distributed lag model for an increase in PNC of 10$^4$ cm$^3$. Since there is no reason to choose one set of numbers
over another and all these numbers provide a different estimate, we have made separate estimates for each set of relative risk values.

Table S.4. Summary of data used for total mortality estimates (RR denotes relative risk).

<table>
<thead>
<tr>
<th>Items</th>
<th>Details</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Population of Delhi</td>
<td>22.16 million in 2010</td>
<td>[51]</td>
</tr>
<tr>
<td></td>
<td>30.87 million in 2030 (projected)</td>
<td></td>
</tr>
<tr>
<td>Mortality rate</td>
<td>6.29 per 10^3 in 2008 (Annual Report of Registration of Births and Deaths)</td>
<td>[52]</td>
</tr>
<tr>
<td>RR for lag 1</td>
<td>1.013 (1.005–1.022) per 10^4 PNC</td>
<td>[48]</td>
</tr>
<tr>
<td>RR for lag 4</td>
<td>1.030 (1.003–1.056) per 10^4 PNC</td>
<td>[49]</td>
</tr>
<tr>
<td>RR for lag 4 (polynomial distributed lag model, pdl)</td>
<td>1.043 (1.014–1.072) per 10^4 PNC</td>
<td>[49]</td>
</tr>
</tbody>
</table>

S.6 References


11. CPCB, Overview of transport sector in India. *Central Pollution Control Division* 2005, Urban Pollution Control Division, [www.cpcb.nic.in/divisionsofheadoffice/upcd/Profiles.pdf](http://www.cpcb.nic.in/divisionsofheadoffice/upcd/Profiles.pdf)


