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Preliminary estimates of nanoparticle number emissions from road vehicles in megacity Delhi and associated health impacts

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Abstract. Rapid urbanisation in developing megacities like Delhi has resulted in an increased number of road vehicles and hence total particle number (ToN) emissions. For the first time, this study presents preliminary estimates of ToN emissions from road vehicles, roadside and ambient ToN concentrations, and exposure related excess deaths in Delhi in current and two future scenarios; business as usual

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(BAU) and best estimate scenario (BES). Annual ToN emissions are estimated as 1.37×10^{25} for 2010 which are expected to increase by ~4 times in 2030–BAU, but to decrease by ~18 times in 2030–BES. Such reduction is anticipated due to a larger number of compressed natural gas driven vehicles and assumed retrofitting of diesel particulate filters to all diesel vehicles by 2020. Heavy duty vehicles emit the majority (~65%) of ToN for only ~4% of total vehicle kilometres travelled in 2010. Their contribution remains dominant under both scenarios in 2030, clearly requiring major mitigation efforts. Roadside and ambient ToN concentrations were up to a factor of 30 and 3 higher to those found in respective European environments. Exposure to ambient concentrations resulted in ~508, 1888 and 31 mortalities per million people in 2010, 2030–BAU and 2030–BES, respectively.

1. Introduction

Rapid urbanisation has resulted in a considerably increased number of road vehicles in megacities over the past few decades, making their inhabitants vulnerable to air pollution induced health risks [1]. Atmospheric nanoparticles are one of the air pollutants which are currently not regulated through air quality standards in any developing or developed megacities. Up to ~85% of total particle number (ToN) concentrations in polluted urban environments originates from road vehicles [2]. More than 80% of ToN concentrations in atmospheric urban environments reside in the ultrafine size range (i.e. <100 nm in diameter) that contribute almost negligibly to particle mass concentrations [3]. The particle size range below 300 nm (referred here as nanoparticles) constitute over 99% of ToN concentrations in urban environments [2]. Therefore, in what follows, the terms ToN and nanoparticles are used interchangeably as are the terms ambient, airborne and atmospheric (according to the context). New sources such as manufactured nanomaterials [4] have recently emerged but road vehicles remain the largest contributors to the ToN emissions [5]. The vehicle population in developing megacities like Delhi is expected to increase substantially in future years. This means an increased level of ToN release into the urban atmospheric environment resulting in adverse effect on human health, urban visibility and global climate [2, 6].

Currently, there are no air quality standards in any part of the world to limit public exposure of atmospheric particles on a *number* basis since current regulations are based on mass concentrations of PM_{10} ($D_p \leq 10$ μm) and $PM_{2.5}$ ($D_p \leq 2.5$ μm) [5]. Recent inclusion of particle *number* emission limits for vehicles in Euro–5 and Euro–6 standards for light duty diesel vehicles is the first ever initiative to control them at source in European countries [2]. Stricter emission standards, cleaner fuels, advances in engine and after–treatment emission technologies and introduction of cleaner (hybrid) vehicles have significantly reduced emissions of particulate mass and gaseous pollutants in developed urban cities [5, 7]. However, implementation of such emission policies and control measures may take decades to come in force in developing countries.

Delhi's population in 2010 were about 22.16 million which was distributed over a surface area of 1483 km^2 [8] – this is about 2.6 times larger than the London population dispersed over 0.86 times the surface area of Delhi [8]. This indicates a much higher integrated exposure of Delhi's inhabitants to atmospheric nanoparticles compared with developed megacities. Since nanoparticles exposure is often positively related with respiratory and cardiovascular diseases and increased rates of mortality [6], a large number of morbidity and mortality cases can be attributed to nanoparticles which have not been quantified for Delhi until now.

So far, only a small number of emission inventories for fine particulate matter have been constructed. Most of these have been for the UK [15-17] or Australia [9] but none of them corresponds to a developing country. Moreover, these inventories restrict their scope to estimation of PM_{10} and $PM_{2.5}$ emissions, except Keogh et al [9], who recently published a comprehensive emission inventory for urban South–East Queensland in Australia considering both particulate mass (PM_{10} and $PM_{2.5}$) and numbers. For the first time, our study makes *preliminary* estimates of ToN emissions, roadside and ambient ToN concentrations and associated total mortality in Delhi under two future scenarios: business as usual (BAU), and best estimate scenario (BES). We have used the word '*preliminary*' because a number of assumptions are used in estimations due to the lack of location specific data. Also note that our study only focuses on particle *number* emissions only from *road vehicles* in the megacity Delhi; other emission sources are not considered.

2. Methodology

This section briefly presents key information on study area, modelled scenarios, estimates of vehicle types, their population and vehicle kilometre travelled (VKT). A detailed description on the topics covered below can be seen in supplementary Sections S.1 and S.2.

2.1 Description of the study area

Delhi (28°38'17"N, 77°15'51"E) is among the foremost developing megacities in the world. Its inhabitant population increased by 21.5% in 2006 from the 2001 levels compared with 7.5% increase in national population [8]. The population is further expected to increase by about 54% in 2030 from the 2006 levels [8]. Delhi's transport system mainly relies on roads. In 2008, Delhi had about 31,183 km road length with 100's of flyovers [10] which is growing with the ongoing development of a bus rapid transit system (BRT). A total of 26 (7, 3 and 16) BRT corridors are planned in three five–yearly phases starting from 2005; these will cover a total length of 310 km by the year 2020 [11]. The surface area used by roads is about 21% of Delhi's total land area [12], covering about 1749 km of road length per 100 km^2.

Buses are the dominant mode (~42% of total personal trips in 2007–2008) of transportation that is followed by cars, 2–wheelers (2Ws; motorcycles and scooters), 3–wheelers (3Ws; auto–rickshaw) and bicycles [11]. Considerable efforts are being made to reduce air pollution levels in the city by implementing a clean fuel policy and developing transport infrastructure (e.g. BRT and metro). For instance, the majority of vehicles were operating on diesel and gasoline fuels prior to 2001. In 2001, the Delhi government strictly implemented compressed natural gas (CNG) fuel for operation of buses and 3Ws, which was applicable for light duty vehicles (LDVs; those <3.5t in weight) from 2006. Following the orders of the Supreme Court in April 2001, transport such as buses, 3Ws and all commercial vehicles including taxis aged over 15 years were required to be changed to CNG. These orders also included introduction of Euro–I emission standards for private passenger cars (cars and jeeps), use of unleaded petrol, and premixing of 2T (two stroke) oil with petrol for 2Ws.

Delhi is surrounded by two states (Uttar Pradesh and Haryana) and is also a central point for buses to transport the passengers to other states in India. Consequently, a considerable amount of inter–state traffic (mainly diesel–fuelled buses and heavy duty vehicles, HDVs) enters and passes through the city everyday.

2.2 Modelled scenarios

Emission estimates of ToN concentrations are made between 1991 and 2030 but the levels of 2010 are considered as a baseline figure to compare with 2030 estimates in two modelled scenarios (BAU and BES). BAU is a base case scenario in which no policy interventions are considered. Detailed construction of the BAU gives 5.40 and 5.58 times increase in total vehicle population and 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 those 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. Whereas, BES considers promising reduction measures in nanoparticle emissions due to interventions by transport and emission control policies and infrastructural development for road transport. Detailed construction of this scenario results in 3.09 and 4.03 times increase in total vehicle population and VKT, respectively, in 2030 from the 2010 levels. 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 Clean Air Initiative for Asian cities. Detailed methodology describing the construction of these scenarios is presented in Sections S.1 and S.2.

2.3 Modelling ToN emissions

The ToN emissions (# yr^{-1}) are estimated using the Eq. (1) which is a product of PNEF (# veh^{-1} km^{-1}) and VKT by each vehicle type in a year.

$$ToN = (PNEF_{cars} \times VKT_{cars}) + (PNEF_{2w} \times VKT_{2w}) + (PNEF_{3W} \times VKT_{3W}) + (PNEF_{Buses} \times VKT_{Buses}) + (PNEF_{HGVs} \times VKT_{HGVs}) + (PNEF_{LGVs} \times VKT_{LGVs}) -------------- (1)$$

Six vehicle categories are considered for the estimates: passenger cars and jeeps (gasoline, diesel and CNG), 2Ws (gasoline; two and four strokes), 3Ws (gasoline and CNG), Buses (diesel and CNG), LDVs (diesel, petrol and cars, and HDVs (diesel; those >3.5t in weight). Subsequent sub–sections illustrate the details of collected PNEF and VKT data.

2.3.1 PNEFs

There are no PNEF studies available for road vehicles running in Indian or Asian countries. The majority of available studies are either from the European, American or Australian region [9]. To account for a variety of vehicles driven by CNG, diesel or gasoline fuels, an extensive review of PNEF studies published in the last two decades is carried out (Table S.1) and representative PNEFs are selected for our use (see Table S.2). Under both scenarios, PNEFs were selected for individual vehicle types according to their corresponding speeds during the following designated time periods: 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). Different values of PNEFs are chosen under the BES due to the change in fuel types, speeds and retrofitting of diesel particulate trap (DPF), as illustrated in Table S.2 and Section S.2.3.

2.3.2 Modelling vehicle population, speed and fuel types

Modelling of vehicle population in the BAU ($N_{\text{v,BAU}}$) and BES ($N_{\text{v,BES}}$) is required to accurately quantify the annual VKT. Since there is no consolidated database available for this purpose, we have constructed this data after considering the findings of relevant published studies and sensible assumptions using the following equations:

$N_{\text{v,BAU}}$ = Number of registered road vehicles + External vehicles coming in and passing through the city – Phased out old vehicles as described in Section 2.2

$N_{\mathrm{v,BES}} = N_{\mathrm{v,BAU}}$ – Vehicles off the road due to MRTS and Bus Rapid Transit (BRT) corridors in Delhi

Vehicle population between 1991 and 2030 is compiled using the vehicle registration data for past years and applying a growth factor for future years. Firstly, vehicle registration data in Delhi between 1991 and 2006 are used as a base data for vehicle population [13]. Future growth of vehicles was then 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 the 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 higher than those suggested by Murthy et al. [14] for 3Ws (8%), taxis (5%), buses (7%), cars and jeeps (10%), and 2Ws (9.8%) due to consideration of higher GDP growth than anticipated in past years. Detailed procedure for estimating the $N_{\mathrm{v,BAU}}$ and $N_{\mathrm{v,BES}}$ are provided in supplementary Section S.2.1.

2.3.3 Total VKT under both scenarios

For both scenarios, the annual VKT for each vehicle category are estimated by multiplying the VKT per day with the total number of days in a year. The VKT per day were assumed to be 41 (cars), 27 (2Ws), 110 (3Ws), 164 (Buses), 82 (HDVs and taxis) and 110 (LDVs) [14-15]. Total VKTs are then divided into the periods described in Section 2.3.1, i.e. peak (53%), off–peak (40%) and free flow (7%) for choosing vehicle–speed specific PNEFs during these periods. 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) [16]. An increase of 11% from peak hours is considered for off peak hours [16-17]. During the free flow traffic conditions, which usually occur at night, the maximum permissible speed for vehicles was capped at 60 km h^{-1} under both scenarios [18]. Under the BES, average vehicle speed is taken as the vehicle speeds during the BAU plus the increase due to infrastructural development as explained in Sections 2.2 and S.2.2.

2.4 Estimation of total mortality related to changes in ToN concentrations

In order to calculate 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. [6] from a time series study conducted in London, and by Stolzel et al. [19] for Erfurt, Germany. 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. 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 [8] and the mortality rate from the Annual Report on Registrations of Births and Deaths for Delhi [20]. Detailed description of the estimation method and the data used is provided in Section S.5.

3. Results and discussion

3.1 Modelled estimates of ToN emissions

Annual ToN emissions in 2010 are estimated as 1.37×10^{25} which is expected to increase ~4.21 times in 2030–BAU (Table 1). This increase was anticipated since the VKT values increased due to ~5.58 times growth in vehicle population in 2030–BAU compared with 2010 levels. One way to compare the results in chosen scenarios is to normalise the emissions by the VKT values. The emissions to VKT ratio was 1.99×10^{14} km^{-1} in 2010 which slightly decreased to 1.43×10^{14} km^{-1} in 2030–BAU due to the replacement of retired vehicles with the new CNG vehicles in traffic fleet.

Under the 2030–BES, annual ToN emissions decreased by about two orders of magnitude (7.8×10^{23} yr^{-1}) from the 2010 levels. This resulted in about three orders of magnitude smaller emissions to VKT ratio (6.02×10^{11}) than those in 2010 (1.99×10^{14}). The main reasons for these countable reductions were the rapid phasing out of gasoline and diesel driven taxis, buses and LDVs and their replacement with the new CNG vehicles. The other key factor responsible for this reduction was assumed retrofitting of DPFs on all diesel vehicles by 2020 since these can decrease the ToN emissions by about two orders of magnitude or more compared with non–DPF diesel engines [21].

Dividing the ToN emissions by inhabitant population gives per capita per day emissions. This was found to be 1.70×10^{15} in 2010 which increases by about 3 times in the 2030–BAU but decreased substantially (i.e. 243 times) under the 2030–BES due to a favourable combination of both increased inhabitant population and decreased emissions. These observations also indicate that a considerable reduction in the ToN emissions can be achieved if the assumptions considered in the BES are implemented, benefiting both the local air quality and public health (see Section 3.6).

3.2 Contribution of different vehicle types to ToN emissions

Table 2 illustrates the VKT and ToN emissions contributed by different vehicle types. As opposed to the VKT contribution by CNG driven vehicles, the share of diesel and gasoline vehicle driven VKTs decreased in future years due to a favourable shift towards the CNG fuel. Despite this, contribution to ToN emissions from all diesel vehicles remains dominant in both future scenarios; emissions from gasoline and CNG vehicles follow. The BES targets the largest contributor to ToN emissions (i.e. diesel vehicles) and brings about 34 times decrease in 2030–BES from the 2010 levels and an increase in CNG and gasoline contributions to about 13 and 2 times, respectively (Table 2).

If we look at the different vehicle categories in 2010 and 2030–BAU, passenger cars (taxis, cars and jeeps) are the highest contributor to the VKTs, followed by the 2Ws, buses, 3Ws, HDVs and LDVs. Passenger cars are however the second largest contributor (25–34%) to ToN emissions after the HDVs in all scenarios, mainly due to their larger population running on gasoline and diesel fuel. Contributions of 2Ws towards the VKTs are second largest (35–39%) but they contribute substantially less (0.26–0.38%) to the ToN emissions in 2010 and 2030–BAU. One of the findings in accordance with a recently published study [9] is the contribution of the HDVs to the ToN emissions. The HDVs contributed 4.26% of total VKT in 2010 but they alone emitted ~65% of ToN emissions. Consistent with this were the observations in 2030–BAU and 2030–BES where the HDVs added to ~2.59 and 4.89% of total VKT, but corresponded to ~52 and 51% of ToN emissions, respectively. The HDV population is expected to be tripled (3.39 times) in 2030 over the 2010 values under both scenarios, suggesting that emissions control from the HDVs require major mitigation efforts in future. Contribution to ToN emissions from the HDVs remain dominant even when the after–treatment systems (i.e. DPF) are assumed to be used under the BES. One of the predominant reasons for the HDVs to be the largest contributor is their much larger PNEFs compared with other vehicles (see Table S.2). This is presumably a leading explanation that our estimates of annual ToN emissions (1.37×10^{25}) compared well with those estimated (1.08×10^{25}) by Keogh et al. [9] for South–East Queensland in Australia. The HDVs contributed to about 54% of their annual ToN emissions although they added only 6% to total VKT.

3.3 Estimating ToN concentrations

Equation (2), which is based on a simplified box model (see Section S.3 for detailed formulation), is used to convert the annual ToN emissions into the hourly averaged *roadside* and *ambient* ToN concentrations:

$$\text{Hourly averaged ToN concentrations (\# cm}^{-3}\text{)} \approx \frac{Q\times L}{H_m\times U_r}=\frac{ToN\times L}{A_s\times H_m\times U_r} \qquad (2)$$

where 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. Two different values of A_s are considered for mimicking the ambient (~15m) and roadside (~2m) concentrations. Detailed description of data used for these estimates are provided in Section S.3.1.

The resultant ToN concentrations from the Eq. (2) are presented in Table 3. It is worth noting that these concentrations are *derived from the road vehicles* only. The contribution from other sources (e.g. background, light petroleum gas, wood and biomass burning for cooking, small–scale industries, power plants and exhaust–emissions from non–road construction machinery) can not be neglected while speculating upon the total ToN population in Delhi's ambient environment [22-23]. A recent source apportionment study for Barcelona city found about 35% of total ToN emissions from other sources [24] but such contributions are largely unknown for Delhi and are expected to be much larger [22-23]. Our ambient ToN concentrations are still up to 3 times larger compared with overall concentrations in the ambient urban environments of European [25] or American [26] cities. If we compare the vehicle–derived component of our ambient ToN concentrations (3.27×10^4 cm^{-3}), these were ~3 times higher to those observed (1.14×10^4 cm^{-3}) by Pey et al. [24] in the ambient environment of Barcelona as a contribution from road vehicles. Furthermore, our ambient ToN concentrations compare well with a unique study for Delhi by Monkkonen et al. [23]. In 2002, they measured ToN concentrations in the 3–800 nm range at a height of 15 m and close to a traffic lane in a residential area adjacent to India Habitat centre (New Delhi). They found highest measured 24–h average concentrations in the range of $(6.28\pm1.78)\times10^4$ cm^{-3}, with the lowest and highest concentrations being 2×10^4 and 2.5×10^5 cm^{-3}, respectively. We mimicked our ambient ToN concentrations to the 2002 levels for making a comparison. As expected, our estimates, $3.17(2.02–7.33)\times10^4$ cm^{-3}, are at the lower end of the concentrations measured by Monkkonen et al. [23] since these exclude contributions from above–described sources.

Our *roadside* ToN concentrations are generally about a factor of 23 times larger than our *ambient* ToN concentrations. These turns out to be about 23, 26 and 29 times larger than those found along the roadsides in London, UK [27], Stockholm, Sweden [36] and Cambridge, UK [3], respectively. Roadside ToN concentrations are expected to grow about 4–fold in 2030–BAU, but about 18–fold decrease in 2030–BES, from the 2010 levels (Table 3). The 2030–BES remarkably bring down both the ambient and roadside ToN concentrations to well below the corresponding current levels found in a developed megacity like London [28].

3.4 Effects of transformation processes on estimated ToN concentrations

Health impacts are quantified due to exposure of *ambient* ToN concentrations (Section 3.5). Separate estimates are not made for the *roadside concentrations* because of the unavailability of population exposure data along the roadsides in Delhi. To avoid chances of extreme health impact estimates, the *ambient* ToN concentrations are corrected for the possible losses due to transformation processes such as dry deposition, coagulation and nucleation since these can have a substantial effect [29] in scenarios (e.g. 2030–BAU) with high ToN concentrations. Other processes like condensation and evaporation are ignored due to the following reasons. These are reversely acting simultaneous processes and partly negate each other's effect and condensation does not affect ToN concentrations [2]. Majority of evaporation occurs to the nucleation mode liquid particles immediately after their formation near the tail pipe by nucleation and condensation during initial dilution and cooling [30]. A recent study by Dall'Osto et al. [31] for London found that evaporation is substantially important to remove the sub–30 nm particles on distance scales of the order of 1 km and travel times of around 5 minutes upon moving away from major sources. Since ambient ToN concentrations used for health impacts analysis in our study are estimated at about 15 m height above the ground level, our distance and time scales to reach to this height are much smaller than those suggested by Dall'Osto et al. [31]. There could be a small increase (~1% of tailpipe emissions; Dahl et al., [32]) in ToN concentrations due to the particles generated by the road–tyre interaction and brake wear which is also neglected.

Our estimated ambient ToN concentrations do not provide information on the size distributions which is required for making loss estimates due to coagulation and dry deposition. Therefore, we have adopted the particle size distributions which were measured by Monkkonen et al. [23] for Delhi. They found geometrical mean diameters (GMD) in nucleation, Aitken and accumulation modes as ~11, 44 and 147 nm, respectively, with distributions of ToN concentration in these modes as ~8, 58 and 34%, respectively (Table S.3). For approximating the losses, coagulation coefficients and dry deposition velocities for these GMDs were estimated by assuming monodisperse distributions in each mode (see Section S.4). Formation rate of 3 nm particles were found to be varying between 3.3 and 13.9 cm^{-3} s^{-1} in Delhi's environment [23], which represent typical formation rates of new particles in urban conditions [33]. We used 3.3 cm^{-3} s^{-1} for making a *conservative* estimate for the production of new particles due to nucleation. Percent changes in ToN concentrations due to coagulation, condensation and nucleation in different scenarios are illustrated in Table S.4, and corrected ToN concentrations in ambient environment of Delhi are shown in Table 3.

As expected, coagulation losses were highest (~13% of ToN concentrations) due to the largest ToN concentrations in 2030–BAU (Table 3) compared with ~4 and ~0.2% in 2010 and 2030–BES, respectively. Dry deposition losses were about ~11% in all cases. Formation of new particles due to nucleation was highest for the 2030–BAU (~3% of ToN concentrations), followed by negligible contributions in 2010 (~0.2%) and 2030–BES (~0.1%) which is expected due to a large condensation sink and background particle loading in Delhi. The net losses during all scenarios ranged between 10 and 22% compared with inert treatment of particles (Table S.4). These losses are identical with the detailed modelling studies of Ketzel and Berkowicz [34-35] for Copenhagen city where they found net losses between 10 and 30%, and of Gidhagen et al. [36] for Stockholm city where they found coagulation and dry deposition losses up to 3 and 25%, respectively.

3.5 ToN exposure in megacity Delhi and in typical urban locations

Exposure to high ToN concentrations may aggravate existing pulmonary and cardiovascular diseases due to efficient alveolar deposition of tiny particles and their potential to enter the pulmonary vascular space [37-38]. Fresh vehicular exhaust contains many nanosized particles that take a few seconds of travelling time to reach to the roadside [39] where people living, walking or travelling by motor vehicles, bicycles and 2Ws are exposed [40]. Concentration levels of exposure can vary up to two orders of magnitude or more depending on the exposed location. For instance, concentration levels for exposure can be to ~10^6 cm^{-3} while travelling in car in urban or tunnel routes [41], ~10^5 cm^{-3} during cycling, walking or travelling in buses in heavily trafficked area [40, 42], and ~10^4 cm^{-3} in typical street canyon conditions [43]. Our estimated roadside concentrations in 2005 were 6.95(4.43–16.08)×10^5 cm^{-3}; these are about 6–10 times larger than those measured by Kaur et al. [42] at *a heavily trafficked route* in London during their exposure assessment study for the people walking (0.68×10^5 cm^{-3}), cycling (0.94×10^5 cm^{-3}), travelling in buses (1.01×10^5 cm^{-3}), cars (0.99×10^5 cm^{-3}) and taxis (0.88×10^5 cm^{-3}).

For Delhi inhabitants, current exposure to ambient and road side concentrations are of the order of ~10^4 and ~10^5 cm^{-3}, respectively (Table 3).

3.5.1 Total mortality due to exposure of ambient ToN concentrations

Using the methodology described in Section 2.4, estimates of deaths brought forward (total mortality) are made due to the exposure of *corrected ambient* ToN concentrations in different scenarios using the exposure model of Atkinson et al. [6] for a 1 day lag and Stölzel et al. [19] for a 4 day lag (see Table 3, and Section S.5 for model details). Consistently lowest estimates are produced by the model of Atkinson et al. [6] while the largest derive from the polynomial distributed lag (pdl) model of Stölzel et al. [19]. All the models show a large uncertainty which is reflected in the long 95% confidence interval (CI) range in Table 3. Inter–comparison of average mortalities derived from different models indicates a factor of 1.42 to 3.31 differences. This is evident from the following averaged mortalities over all the modelled results in each scenario which are used for further discussions: 11252(95% CI=2872–19580), 58268(14871–101394), and 952(243–1657) in 2010, 2030–BAU and 2030–BES, respectively.

Total mortality attributable to nanoparticle exposure in 2010 is anticipated to increase to about 5 times in 2030–BAU. Because of much lower ambient ToN concentrations (Table 3) in the 2030–BES, total mortalities are expected to decrease about 12 and 61 fold compared with 2010 and 2030–BAU levels, respectively. Our mortality figures should be interpreted as 'lower estimates' since these are based on the corrected ambient ToN concentrations, much lower than those expected at a breathing height of ~2 m, and are derived from road–vehicles only. We have chosen ambient ToN concentrations for mortality estimates because these are most relevant to exposure of the entire Delhi population, including the people living in high rise buildings.

These are the first ever mortality estimates associated with nanoparticle exposure for Delhi. In fact, no such mortality figures are currently available for a megacity in any part of the world. This also strips the opportunity to directly compare our estimates with the published literature. Therefore, we have selected few Delhi specific studies, which have made mortality estimates for other air pollutants, for discussing the relative health impact of so far overlooked nanoparticles. For instance, a recent study by Gurjar et al

[1] estimated mortalities due to exposure of air pollutants for a number of megacities, including Delhi. They found the total mortality due to nitrogen dioxide (NO_2) and total suspended particulate matter (TSP) exposure as 167 and 11424, respectively, for the year 2005 (i.e. 10 and 680 mortalities per million people for NO_2 and TSP, respectively). For the same year in Delhi, a recent study estimated total number of cardiopulmonary related deaths between 1700 and 2600 for the people aged *over 30 years* due to $PM_{2.5}$ exposure; these gives an average mortality for this age group as 251 per million people [44]. We mimicked our mortality estimates to 2005 levels for comparing the mortalities due to other air pollutants in Delhi. These turned out to be 7943 (i.e. 473 mortality per million people) based on corrected ambient ToN concentrations in 2005. Normalisation of above mortalities figures provides an approximately 0.69, 1.88 and 48 times relative mortality impact by *vehicle–derived* nanoparticles in Delhi than those by all sources derived TSP, $PM_{2.5}$ and NO_2, respectively. These normalised figures should not be seen as a general impact of nanoparticle related mortalities compared with other air pollutants since concentrations of nanoparticles and other air pollutants can vary in different cities depending on the types of emission sources, geographical and meteorological conditions, and so will be their relative impact on total mortalities. However, the above discussions clearly indicate that exposure to nanoparticles leads to a considerable number of excess deaths in Delhi which has never been accounted before. Furthermore, a countable increase in total mortalities is expected in future years (e.g. 1888 per million people in 2030–BAU), indicating a serious need to control the nanoparticle emissions at source by considering associated mitigation measures. Total mortalities under the 2030–BES turns out to be modest (i.e. 31 per million people) as a consequence of considered assumptions, mainly the use of DPF for diesel vehicles.

4. Synthesis and future research challenges

This study presents the first published preliminary estimates of road vehicles derived ToN emissions and concentrations in the roadside and ambient environments of megacity Delhi. Total mortalities due to exposure of ambient ToN concentrations to Delhi inhabitants are also made for the first time. All these estimates are made under the current and future years in two different scenarios

(BAU and BES). The study also identifies predominant source of nanoparticle emissions in the Delhi traffic fleet, besides suggesting possible measures through the BES for mitigating their impacts on public health and the environment.

Passenger cars contribute to the largest VKT in all scenarios but their contribution to the ToN emissions was second to the HDVs which emit more than half of the ToN emissions for only ~5% VKT. From the 2010 levels, ToN emissions are expected to increase ~4 times in 2030–BAU compared with ~18 times reduction in 2030–BES, mainly due to assumed implementation of emission control technology (DPF in all diesel vehicles) and greater use of clean fuels (CNG) in future years. Future developments of public infrastructure (MRTS and BRT) modestly influence the results of our studied scenarios. This is mainly because of a marginal increase in the vehicles speed due to decongestion on roads, leading to a negligible change in applied PNEFs and the ToN estimates.

The annual ToN emissions were found to be 1.37, 5.77 and 0.078 ($\times 10^{25}$), and corresponded to ~10^5, ~10^6 and ~10^4 # cm^{-3} roadside concentrations, in 2010, 2030–BAU and 2030–BES, respectively. The ambient ToN concentrations were about 23 times smaller than those found at roadside, and corresponded to about 508, 1888 and 31 mortalities per million people, in 2010, 2030–BAU and 2030–BES, respectively. Because of a peculiar combination of densely populated inhabitants and high ToN concentrations, health impacts related to nanoparticle exposure are expected to be much greater in Delhi than in any developed megacity. Diminishing emissions from the HDVs have appeared as one of the most imperative mitigation strategies for limiting nanoparticle exposure to Delhi public.

The study also revealed several difficulties to carry out such investigation. First and foremost is the lack of location specific data (e.g. PNEFs, relative–risks) which are crucial for imputing ToN emissions and mortalities. This has prompted us to use the word ‘preliminary’ in the title. Although there is no obvious reason to suspect our results as estimates are justifiable and compare well to infrequent studies on this topic (see Section 3). Moreover, the study develops novel methodologies to back–calculate ambient and roadside ToN concentrations, and associated total mortalities. Concepts of these methodologies are transferable to any developing megacity where measurements of nanoparticles are scarce and health impacts due to nanoparticles exposure have rarely been assessed. Evaluation of emissions and health impacts in different scenarios also provide a sound basis for the local regulatory authorities to assess the future ToN emissions and accordingly design mitigation strategies for limiting their impact on public health and the environment.

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6. Supporting information

Please see Sections S.1–S.5.

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Table 1. Summary of nanoparticle estimates in various scenarios.

Year	Inhabitant population ($\times 10^6$)[a]	Vehicle population ($\times 10^6$)	Total VKT per annum ($\times 10^{10}$)	ToN per annum ($\times 10^{25}$)	Emissions per person per day ($\times 10^{15}$)
2010	22.16	4.74	6.91	1.37	1.70
2030 (BAU)	30.87	25.6	38.56	5.77	5.13
2030 (BES)	30.87	9.26	20.41	0.078	0.007

[a] Delhi's population is extrapolated to 2030 from the predictions given by the World Health Organisation [8] for 2015 (24.16 million), 2020 (26.27 million) and 2025 (28.57 million).

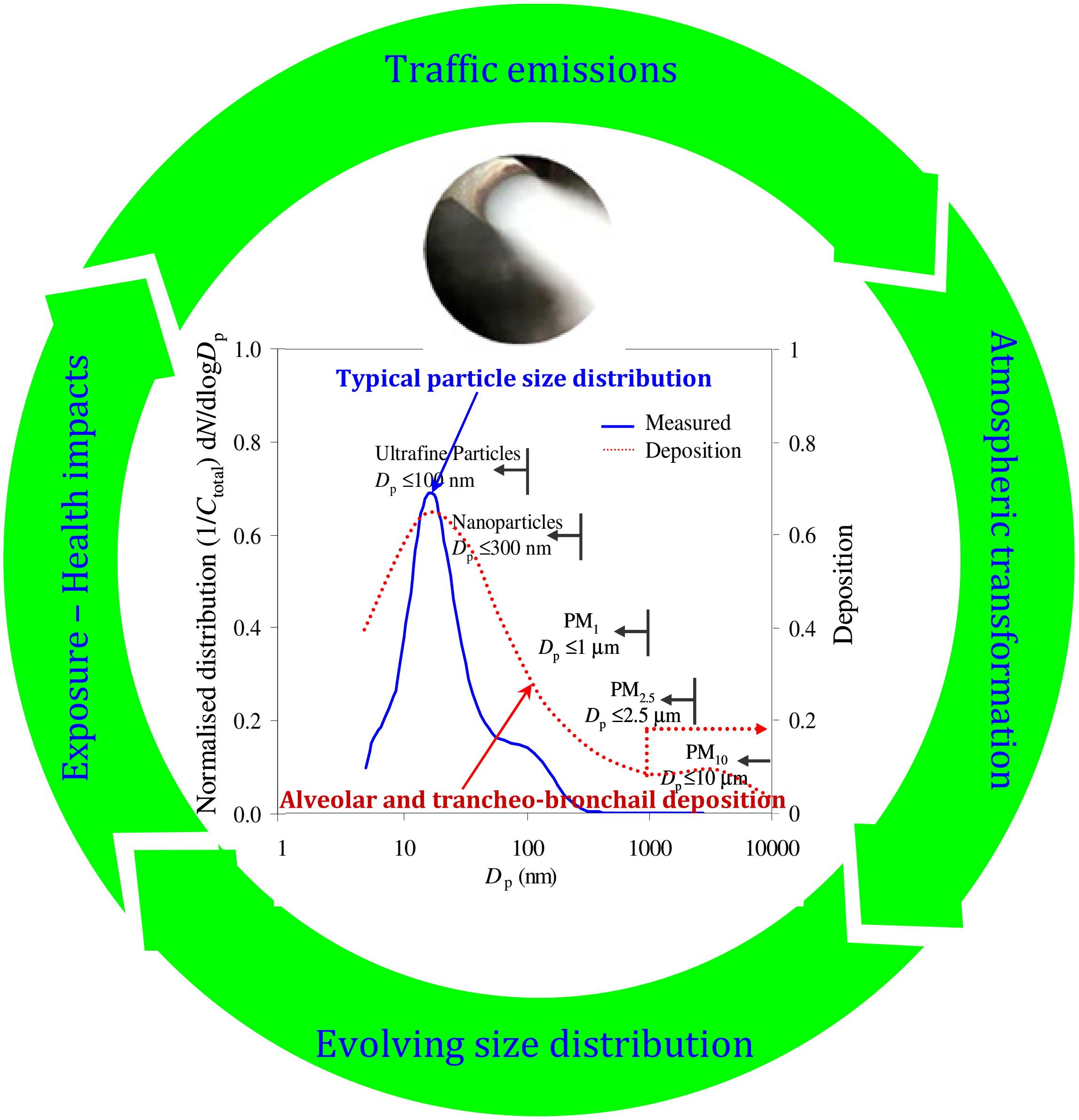
Table 2. Contribution of different vehicle types towards total VKT and ToN emissions.

All vehicle types	ToN emissions ($\times 10^{23}$)			VKT contribution (%)			ToN emission contributions (%)			VKT (% change from 2010)		ToN emissions (% change from 2010)	
	2010	2030 (BAU)	2030 (BES)	2010	2030 (BAU)	2030 (BES)	2010	2030 (BAU)	2030 (BES)	2030 (BAU)	2030 (BES)	2030 (BAU)	2030 (BES)
Cars and Jeeps	34.96	148.99	2.64	41.09	31.30	36.57	25.44	25.8	33.74	325	163	326	-92
2W	0.36	2.22	0.27	34.81	38.41	9.03	0.26	0.38	3.53	516	-52	516	-23
3W	0.02	0.10	0.09	5.32	6.68	11.13	0.02	0.02	1.15	601	39	326	271
Taxi	0.62	1.66	0.25	2.07	1.08	1.7	0.45	0.29	3.14	192	11	170	-60
Buses	7.59	99.14	0.52	7.25	15.70	28.67	5.52	17.17	6.7	1109	5	1206	-93
HDVs	89.13	302.01	4.03	4.26	2.59	4.89	64.85	52.29	51.52	239	239	239	-95
LDVs	4.75	23.42	0.02	5.2	4.24	8.01	3.46	4.06	0.23	355	-18	393	-100
All diesel	135.83	570.10	4.07	46.54	39.35	35.90	98.83	98.71	52.06	372	128	320	-97
All gasoline	1.56	6.99	3.11	42.25	43.69	14.98	1.14	1.21	39.81	477	5	347	99
All CNG	0.04	0.45	0.64	11.21	16.96	49.12	0.03	0.08	8.13	744	1194	878	1277

Table 3. Averaged ambient and roadside ToN concentrations in different scenarios; figures in parenthesis represent standard deviation related lower and upper values of concentrations. Excess deaths are derived from the ambient ToN concentrations (after losses) and figures in parenthesis are 95% CI values.

Year	ToN concentrations (×10^4 # cm^{-3})			Excess deaths (total mortality)		
	Ambient		Roadside	Atkinson et al. [7] – lag 1	Stolzel et al. [25] – lag 4	Stolzel et al. [25] – lag 4 (pdl model)
	Estimated	After losses	Estimated			
2010	3.27 [2.08–7.56]	2.81 [1.82–6.17]	74.60 [47.58–172.60]	5091 [1958–8615]	11826 [1175–21930]	16839 [5482–28195]
2030 (BAU)	13.73 [8.76–31.78]	10.44 [7.05–20.05]	311.23 [198.50–720.30]	26362 [10139–44613]	61242 [6084–113561]	87199 [28390–146007]
2030 (BES)	0.19 [0.12–0.43]	0.17 [0.11–0.39]	4.21 [2.69–9.75]	431 [166–729]	1001 [99–1856]	1425 [464–2387]

TOC GRAPHIC



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