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New Directions: Air pollution challenges for developing megacities like Delhi

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Manuscript text

Most major cities around the world experience periods of elevated air pollution levels, which exceed international health-based air quality standards (Kumar et al., 2013). Although it is a global problem, some of the highest air pollution levels are found in rapidly expanding cities in India and China. The sources, emissions, transformations and broad effects of meteorology on air pollution are reasonably well accounted in air quality control strategies in many developed cities; however these key factors remain poorly constrained in the growing cities of countries with emerging economies. We focus here on Delhi, one of the largest global population centres, which faces particular air pollution challenges, now and in the future.

In 1970 there were eight megacities (population >10 millions) worldwide, which increased to 32 in 2015, with a total population of 620.4 million (City Population, 2015). Delhi and its National Capital Region (NCR; Figure 1a) ranked fifth among these megacities, with a population of 25.8 million (SI Figure S1). There were 4.74 million road vehicles in Delhi in 2010 and that number is predicted to increase to 25.6 million by 2030 (Kumar et al., 2011). This increase has major consequences for energy consumption, and hence, emission of air pollutants. For example, total energy consumption from stationary and mobile sources in Delhi had grown by 57.16% from 2001 levels to 230,222 TJ in 2011 (Kennedy et al., 2015). There are no reliable sources available to give corresponding values for energy consumption in the NCR of Delhi. However, a comparable increase in energy consumption can be expected, given a population growth of about 25% in 18 NCR cities (i.e. 9, 7 and 2 in Haryana, Rajasthan and Uttar Pradesh states, respectively) in 2011 compared with ~21% in Delhi from the 2001 levels (Regional Plan, 2021). These surrounding states in NCR follow Central Pollution Control Board (CPCB) standards or evolve their own standards which should be stringent than the CPCB standards for *pollution control*, but use their own norms for *fuel quality* and *vehicle registration* that are usually lenient to those in Delhi.

There are many distinctive features in Delhi which impact on air pollution: (a) uncontrolled sources in its *surrounding perimeter* (where city regulations do not apply and those that apply are not followed stringently), (b) a number of unregulated and unaccounted sources *within the city*, and (c) unfavourable geographic location and regional meteorology, with windy and dusty conditions during summer that are (i)

exacerbated by low relative humidity enhancing particle resuspension, (ii) by episodic dust transport events from surrounding areas (Guttikunda and Gurjar, 2012), (iii) input of ozone as regional background, and (iv) influence of long-range transport of precursor emissions from both the subcontinent and intercontinental sources (West et al., 2009). The deterioration of air quality is further exacerbated by the diurnal cycle of “trucks” which are allowed to pass through the city after about 10 pm (local time) when the height of the mixing layer is also relatively low, due to cold northern and north-easterly winds passing through the city (Guttikunda and Goel, 2013).

The geography of Delhi results in the advection of air into the city from the surrounding areas, which can sometimes be significantly more polluted than the city centre itself. As a land-locked megacity, there are limited avenues for the flushing of polluted air out of the city, or its replacement with air from relatively unpolluted marine regions, which means that atmospheric transport from all directions, is likely to add to inner-city pollution. Such features are common in many growing non-coastal megacities (e.g. Tehran, Cairo), where urban growth is increasingly heterogeneous, sometimes unplanned, and is generally at its greatest in the peripheral areas, given the already saturated development of housing and commercial space within the core of these cities. In contrast, those megacities which are located close to coastlines benefit from *sea breezes* that can exchange maritime and urban air on a diurnal cycle. Therefore, cities surrounded by a densely built environment (e.g. Delhi, Mexico City) are at a clear disadvantage in terms of air pollution, which is made worse if those regions have unregulated emission sources, and/or they are semi-isolated by topographical features (e.g. Santiago).

Air pollutants are typically classified as primary (directly emitted) or secondary (those formed in the atmosphere). While secondary air pollutants (such as ozone, volatile organic compounds and nitrogen dioxide) are major contributors to pollution in cities such as Delhi, effective mitigation of these pollutants requires scientific understanding of their precursors and formation processes. Recent measurements in Delhi during winter show that organic carbon and ions (e.g. nitrate and sulphate) contributed about 37 and 20% of the mean PM_{2.5} mass concentrations, respectively (Pant et al., 2015). Much of the organic carbon and the inorganic components such as nitrate would be expected to be secondary in origin, although both the precise budget and precursor source identities require definitive confirmation. The purpose of this work, however,

is to highlight the critical importance of unregulated and unaccounted *primary* sources of pollutants within and around the periphery of developing megacities. Here we focus on the unique air pollution features of land-locked megacities, such as Delhi, and present a discussion of the challenges associated with developing effective control strategies, and identify corresponding directions for future research requirements.

How important and well understood are unregulated and unaccounted sources? A diverse range of pollution sources co-exist in urban environments and comprehensive knowledge of these sources (both location and strength) is central to developing abatement measures, as well as for creating forecasting and prediction systems. While knowledge of the emission characteristics of key conventional sources in the developed world is reasonably good, there is a lack of high quality, spatially disaggregated primary emissions inventories for growing megacities like Delhi, a natural consequence of its fast rate of change in activity and economic growth. This leads to inadequate or rapidly outdated emission factors and the absence of unaccounted and unregulated sources from inventories. Source apportionment (Pant and Harrison, 2012) and emission inventory (Goel and Guttikunda, 2015; Guttikunda and Goel, 2013; Kumar et al., 2011) studies regularly include primary emissions from road vehicles, power plants and particulate re-suspension, even in developing megacities. However, city specific sources are rarely characterised in terms of their location, magnitude, frequency of operation and emission characteristics. Examples for Delhi include brick kilns that use raw wood, agricultural waste or poor quality coal as a fuel, the roadside burning of organic and plastic waste and the unintentional burning of municipal solid waste at landfills, and construction activities (Figure 1). Although information is unavailable, the electronic waste could be a contributor to the release of nanomaterials to the environment (Kumar et al., 2010) in addition to the emissions when it is burned with the solid waste. For example, local emission inventories point to about 5,300 and 7,550 tons yr⁻¹ of PM₁₀ and PM_{2.5} release from waste burning in Delhi, respectively, while the corresponding emissions from construction are 3,250 and 10,750 tons yr⁻¹ (Guttikunda and Goel, 2013).

Another important and poorly quantified source of air pollution in Delhi is *cooking* (Gargava et al., 2014), especially in peripheral areas (Figure 1a), where it is carried out by the burning of solid biomass or cow dung, emitting a mix of gaseous and particulate pollutants. The black colour of “Chulha (cook stove)” provides very direct

Figure 1. Map of Delhi showing (a) surrounding states, (b) location of upwind power plants and population density in Delhi and its surrounding NCR cities in 2011 (Regional Plan, 2021) as well as the unregulated and unaccounted sources such as (c) landfill, (d) brick kiln, (e) loose soil and waste dumping, (f) biomass burning for cooking using chulhas, (g) dry uncovered surface along the roadside, and (h) traffic congestion.

evidence of the incomplete combustion of biomass and soot/organic carbon emissions (Figure 1f). Pant et al. (2015) found a large biomass burning component of PM_{2.5} in samples collected in Delhi in winter, alongside the expected contributions from road traffic, crustal material and secondary nitrates and sulphates, which were likely attributed to a combination of domestic cooking and heating, and off-grid power generation (small generators). Less expected was a high concentration of ammonium chloride, a constituent once prevalent in the European atmosphere, but now rarely seen (Allen et al., 1989). It arises from the neutralisation of hydrogen chloride gas, thought to originate mainly from coal combustion and refuse incineration, by ammonia. In the case of Delhi, this is most likely to arise from regional agricultural emissions outside the city, and hence, is further evidence of the impact of external pollution sources on air quality in the city centre. Like ammonium nitrate, this salt is semi-volatile and summer concentrations of both were found to decrease as a result of the higher air temperatures (Pant et al., 2015).

The use of *diesel generators* for temporary power generation have been considered in some of Indian emission inventories, such as the Six Cities Programme of the Central Pollution Control Board in India (NSR, 2010). Their contributions become increasingly significant in locations with poor resilience or undersupply in the power networks. Diesel generator sources can become particularly important in Delhi during periods of supply vulnerability as a result of extreme summer and winter weather conditions (Kumar and Saroj, 2014). Such climatic effects force the use of invertors (generally for small houses or shops) and make portable generators a key temporary means of electricity supply. Telecommunication towers are another important user base for diesel generators that is again linked with the undersupply and intermittency in the power networks. Data on the number of telecommunication towers, duration of their use through diesel generators or emissions are not readily available. These generators emit pollutants such as nitrogen oxides, carbon monoxide, hydrocarbons, particulate matter and visible smoke that are regulated by the Ministry of Environment and Forests in India through the emission limits for diesel engines below and above 800 kW in generator sets (see SI Table S1). In general, they are run on inferior quality diesel with high sulphur content, serviced sporadically or not at all and there are hardly any effective regulatory checks in relation to their emissions.

Traffic congestion is a common source of pollution in all cities, but is particularly enhanced in Delhi due to very high density of activities and limited infrastructure. Emissions are particularly high at the many traffic intersections within and around the NCR, which is made worse by poor exhaust control measures and the highly heterogeneous nature of the fleet (Figure 1h). Studies have shown in excess of a 20-fold increase in particle number concentrations at traffic lights during decelerating, accelerating or idling conditions compared with free flowing traffic conditions (Goel and Kumar, 2015). Traffic related line-sources also convert to intense point sources on roads where the delay time is usually measured in minutes. Therefore, traffic emission inventories based on average vehicle speed do not adequately capture this important modulation of vehicle emission levels, without due consideration for congestion.

Unregulated *small industries* use a number of different energy source (e.g. burning of biomass, plastic and crude oil), which are typically unaccounted for in existing inventories. Although many small and household industries moved outside the city to the adjoining areas of Delhi following the implementation of the Indian Supreme Court orders in 1996, they still exist in the NCR region contributing baseline emissions to the city. For example, most of the brick kilns are located outside the Delhi city, but in predominantly upwind sectors (Figure 1b), so they may still be making a significant contribution towards the air pollution load imported to the city. Despite Delhi-NCR having a vehicle density almost 10-times lower than that in Chennai (a city on the coast; i.e. 245 to 2093 veh km⁻¹) and a vehicle population only a little over twice that of Chennai (7.3 to 3.7 million), the average levels of PM_{2.5} were nearly 10-times higher in Delhi (198 to 20 µg m⁻³) (Gupta, 2015), reflecting both the contribution of other sources in Delhi and the lack of a land-sea breeze circulation and stable wintertime meteorology.

How big is an impact from local meteorology and geography? The varying building heights in the densely built-up area of Delhi inhibits breathability of the city, and hence the dispersion of emitted pollutants. This is reflected by only 3.31 m² residential floor area per person in Delhi compared with 21.85 m² in Beijing in 2011 (Kennedy et al., 2015). The mean diurnal range of Delhi's ambient temperature has been decreasing steadily (i.e. from 12.48° in 2001 to 10.34° in 2011) due to urban heat island effects (Mohan and Kandya, 2015), with the possible impact of drawing

outside polluted air into the city centre, especially during the low wind conditions which commonly prevail (SI Figure S2). Whilst other major Indian cities (e.g. Chennai, Mumbai or Kolkata) are situated on the coast and are ventilated by sea breezes, Delhi is surrounded by intense activities, both domestic and industrial, with large sources such as biomass burning and diesel generators in the surrounding NCR region (Figure 1b). Estimating future air quality in Delhi will therefore require both an improved estimation of emissions (from the city itself and from the surrounding regions), as well as representative simulations of the physical transport of pollutants over urban to regional scales.

Are current monitoring and modelling efforts sufficient? In terms of concentration measurements, >20 monitoring stations are run by the CPCB, Delhi Pollution Control Committee and the Indian Institute of Tropical Meteorology Pune. Most of these stations monitor pollutants of regulatory interest such as particulate matter less than 10 μm (PM_{10}) and 2.5 μm ($\text{PM}_{2.5}$), sulphur dioxide, carbon monoxide, ozone and nitrogen oxides. Data in refined forms are accessible to public through the CPCB web portal (<http://www.cpcb.gov.in/CAAQM/mapPage/frmdelhi.aspx?stateID=6>) while raw data can be assessed through requests from users. Monitoring stations running as a part of SAFAR (System of Air quality and weather Forecasting And Research) produce summary data in the form of air quality index (http://safar.tropmet.res.in/index.php?menu_id=1). With two exceptions, these stations are within the Delhi city and coverage of the NCR surrounding Delhi is minimal. This limits observational assessment of the influence of regional pollution advection on air quality in the city itself. Assessment of pollutant levels, and of precursors to secondary species, in the inflowing air from the NCR regions will be vital in determining the causes of poor air quality within the city perimeter, and should be included in developing source apportionment tools to assess the contribution of various sources in business-as-usual and future intervention scenarios (Kumar et al., 2011). A number of models have been adopted for dispersion modelling purposes in Delhi (Gulia et al., 2015). However such models inadequately describe the processing of primary pollutants and the formation of secondary components. This limits their applicability and usefulness in complex built-up environments, such as Delhi, which have unaccounted sources that need to be systemically measured in terms of both their emission levels and the resulting impact on ambient concentrations. The immediate periphery of Delhi is surrounded by two different states (Uttar Pradesh and Haryana;

Figure 1a), both known for their agricultural activities, which contribute significantly to regional pollution (especially at the end of the annual crop cycle when waste biomass is burned in wheat and sugarcane fields), which in turn adds to Delhi's air pollution baseline. A database derived from inflow flux measurements in peripheral areas could assist in evaluating the performance of existing regional scale models or developing new models that are appropriate for application in the local environment. Therefore, the integration of local source apportionment tools, understanding of within-city emissions processing forming secondary pollutants, and city and regional scale dispersion/chemical transport models is necessary to assist in managing the quality of air in Delhi and similar cities elsewhere.

Are interventions and solutions working? The use of after-treatment devices, such as SCR for NO_x reduction and DPF for particulate matter reduction, are effective for reducing emissions from heavy-duty diesel vehicles, which contribute significantly to the emissions in Delhi (Kumar et al., 2011). Further, following the 1998 Directive of the Supreme Court, all public transportation systems in Delhi run on Compressed Natural Gas (CNG), which has substantially reduced visible air pollution as well as the larger fraction of particulate matter. However, issues related to fine particles and other non-exhaust emissions remain unresolved. Whilst the widespread use of flyover bridges in Delhi has had some positive impact in reducing congestion, growth in the demand for transportation, as well as the number of new cars on the roads, limits the effectiveness of flyovers. In addition, many of the surrounding areas, which have not benefited from construction of new flyovers, have inefficient traffic management systems. This means that there are still many vehicles trying to enter/exit the city, particularly during morning and evening rush hours, which run on fuel of inferior quality (e.g. high sulphur content due to lack of uniform fuel quality standards across the country) and adds to on-road congestion (Figure 1h). These may be some of the reasons why, despite emission control strategies, such as the use of clean fuel (e.g. CNG) in all public transport vehicles or the reduction of sulphur content in fuel, the ambient pollution load has not decreased significantly.

In terms of coarse-fraction particulate matter, the resuspension of dust from roads and other unpaved areas (Figure 1c) along with construction activities is a major issue, contributing more than one-third of local PM₁₀ emissions. For example, while road dust has been shown to contribute a significant fraction (~22%) to total PM₁₀

emissions in Delhi in 2010, the corresponding contribution of construction emissions has been about 9% (Guttikunda and Goel, 2013). Dust suppression measures, such as the application of calcium magnesium acetate, magnesium chloride and nano-polymers have been used as localised, temporary pollution reduction measures in European cities (AIRUSE, 2015a). These chemicals are sprayed onto the road surface and bind the particles that it comes into contact with, thereby preventing them from becoming airborne when agitated by the wind, tyre action or vehicle turbulence (AIRUSE, 2015a, b). Positive results have been reported from several case studies (AIRUSE, 2015b), however evidence of the efficacy of such measures is limited and their application is practically and economically unviable on a larger scale. Greening the unpaved roadside areas in Delhi through a natural or artificial grass canopy could possibly help in limiting the resuspension of coarse dust particles during dry and windy seasons. Natural measures, such as the development of wetlands and trees are also effective (Nowak et al., 2013), acting as a sink to remove particulate matter from the air through dry deposition on their surfaces (Tiwary and Kumar, 2014).

What research is needed to address the key issues? A comprehensive understanding of both conventional and unaccounted sources and their emission characteristics is currently lacking. Representative emission inventories detailing the contribution from city- or industry- specific sources are needed. A holistic overview of emissions would also allow the efficient targeting of key individual sources, which if controlled, would lead to maximised benefits. Since the peripheral areas around Delhi are a major source of emissions, an extensive understanding of local versus peripheral, and peripheral versus regional, sources of emissions and their contribution towards local pollutant concentrations is essential. Given the complexity of sources and the influence of meteorology and influx from peripheral areas, Delhi's pollution cannot be managed independently only on a city-wide scale, but requires an integrated approach including the adjacent peripheral regions. This can be realised by, for example, forming urban "city clusters" in Delhi and NCR (Figure 1a), which contribute disproportionately to air pollution load. Understanding the interplay of possible sources and their coordinates within these city clusters could assist in evaluating the effect of any policy, infrastructure or technological interventions on cluster-wise emissions and ambient air pollution concentrations in Delhi and NCR. Such an approach appears to be the most effective means for informing policy, improving air quality and ultimately allowing Delhi to meet international targets for

air quality. Finally, it is important to look beyond monitoring, emission inventories, or source apportionment. Even the best science and technology will not succeed in reducing emissions and improving air quality if it is not considered in a broader framework of economic development of the country, rising awareness of public health risks, and technological progress which is compatible with the nation's cultural, geographical and social context.

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