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Short term association between ambient air pollution and mortality and modification by temperature in five Indian cities

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HIGHLIGHTS

- We model PM_{10}-mortality relationships for five Indian cities across climate zones.
- Higher relative health benefits for pollution reduction in cleaner cities.
- No significant modification effect of temperature on PM_{10}-mortality association.
- The effect observed in this study is similar to those observed in other countries.

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ABSTRACT

Indian cities are among the most polluted areas globally, yet assessments of short term mortality impacts due to pollution have been limited. Furthermore, studies examining temperature – pollution interactions on mortality are largely absent. Addressing this gap remains important in providing research evidence to better link health outcomes and air quality standards for India. Daily all-cause mortality, temperature, humidity and particulate matter less than 10 microns (PM_{10}) data were collected for five cities – Ahmedabad, Bangalore, Hyderabad, Mumbai and Shimla spanning 2005–2012. Poisson regression models were developed to study short term impacts of PM_{10} as well as temperature – pollution interactions on daily all-cause mortality. We find that excess risk of mortality associated with a 10 $\mu g/m^3$ PM_{10} increase is highest for Shimla (1.36%, 95% CI = −0.38%–3.1%) and the least for Ahmedabad (0.16%, 95% CI = −0.31%–0.62%). The corresponding values for Bangalore, Hyderabad and Mumbai are 0.22% (−0.04%–0.49%), 0.85% (0.06%–1.63%) and 0.2% (0.1%–0.3%) respectively. The relative health benefits of reducing pollution are higher for cleaner cities (Shimla) as opposed to dirtier cities (Mumbai). Overall we find that temperature and pollution interactions do not significantly impact mortality for the cities studied. This is one of the first multi-city studies that assess heterogeneity of air pollution impacts and possible modification due to temperature in Indian cities that are spread across climatic regions and topographies. Our findings highlight the need for pursuing stringent pollution control policies in Indian cities to minimize health impacts.

1. Introduction

Short term health impacts of air pollution have been extensively studied for developed countries using time series and case-crossover studies (Lee et al., 2014; Li et al., 2013; Samet et al., 2000; Samoli et al., 2008; Schwartz, 2004). These findings have played an important role in determining air quality standards in the respective countries. For instance, the U.S. Environmental Protection Agency (USEPA) reviews health research every five years to recommend revisions to National Ambient Air Quality Standards, as mandated by the Clean Air Act (Bell et al., 2003; USEPA, 1970). However, epidemiological studies, to inform air pollution policy, are largely limited in the context of developing countries such as India (Balakrishnan et al., 2011).

Indian cities today are among the most polluted areas in the world and it is estimated that outdoor air pollution leads to approximately 670,000 deaths annually (Lim et al., 2013). In India, the Central Pollution Control Board (CPCB) set up under the Air Act of 1981 (MoEF, 1981), is mandated with setting and reviewing the National Ambient Air Quality Standards (NAAQS). Current

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standards, for particulate matter set by the CPCB (CPCB, 2009) are much higher than those recommended by the World Health Organization (Krzyzanowski and Cohen, 2008). In addition, unlike other countries (Bell et al., 2003; Dominici et al., 2007), the CPCB does not take into account findings from health literature when deciding on air quality standards (Balakrishnan et al., 2011). A periodic review of epidemiological evidence informs policy makers about current health risks associated with air pollution and sets the agenda towards finding a balance between reducing health impacts and the costs of implementing further air pollution controls (Dominici et al., 2004).

One potential reason for the lack of tight coupling between ambient air quality standards and health outcomes may be limited epidemiological evidence in the Indian context. A comprehensive review of air pollution and health in Asia found only three time-series studies that examine the short term impacts of air pollution on mortality for the cities of Delhi and Chennai (Balakrishnan et al., 2011; HEI, 2010; Rajarathnam et al., 2011).

However, studies for other cities are needed for at least two important reasons. The first reason is that for a country like India, cities vary widely in terms of development pathways, sources and levels of pollution and policy responses to curb pollution. This presents challenges for generalization of findings from single city studies to the whole country. Second, a changing climate may likely alter pollution levels and subsequently modify health risks over time (Jacob and Winner, 2009; Ren et al., 2006; Tagaris et al., 2009). Consequently, temperature and pollution interactions for cities that lie in different climatic regimes may be quite different. An understanding of these health risks would play an important role in shaping policy to thwart air pollution.

To address the aforementioned research gaps, we use a time-series approach using semi-parametric Poisson regression to study the short term mortality impacts of particulate matter (PM_{10}) as well as temperature – pollution interactions for five cities – Ahmedabad, Bangalore, Hyderabad, Mumbai and Shimla. Being situated in different climactic zones of India, we hope that the observations derived from our findings on these cities will give a fairly good idea about the environment–mortality interaction patterns prevalent in India as a whole.

2. Methods

2.1. Mortality data

Daily all-cause mortality data were collected from the birth and death registers of the municipal corporations of Ahmedabad, Bangalore, Hyderabad, Mumbai and Shimla. For most cities, information on age and cause of death were not available. Table 1 summarizes the climactic characteristics and topography of the above cities.

India is divided into five climate zones namely – hot and dry, warm and humid, composite, temperate and cold. The rationale for choosing these cities was that they are each representative of a different climate zone. In addition to climate zone, these cities represent varied topography – plains, plateau, coastal areas and hilly regions. Air pollution levels vary from city to city based on sources of pollution and policy measures. Additionally, different weather patterns may modify pollution related health risks leading to wide spatial heterogeneity. Thus our choice of cities provides a snapshot of differential health risks across India.

Table 1
Cities distributed by climate zone.

Climate zone	Representative cities	Topography
Hot and dry	Ahmedabad	Plains
Cold	Shimla	Hilly regions
Temperate	Bangalore	Plateau
Composite	Hyderabad, Lucknow	Plains
Warm and humid	Mumbai	Coastal areas

2.2. Weather and PM_{10} data

Daily data on maximum and minimum temperature, relative humidity and dew point temperature were collected from the Indian Meteorological Department (IMD). The IMD has a record of daily weather variables since the year 1948. Daily measurements of PM_{10} were collected from the Central Pollution Control Board (CPCB) database. These included background monitors in residential, industrial and other areas designated as 'sensitive'. Under the National Ambient Air Quality Monitoring Program (NAMP) the CPCB monitors four criteria pollutants i.e. Sulphur Dioxide (SO_2), Oxides of Nitrogen (NO_x), Total suspended particles (TSP) and particulate matter less than 10 microns (PM_{10}) for 342 stations located in 127 cities across India.

Typically two measurements are taken per week for each station implying that 100–120 observations are available per year. These measurements are made available through the CPCB website and the values reported are a 24-hr average. Every city has a different number of air quality monitors that range from one in Shimla to nine in Hyderabad. For a given year, if any monitor had less than 75% of recorded observations (i.e. less than 90 observations), then it was not used in the analysis. Scatterplots of daily mortality, PM_{10} concentrations and temperature for the different cities are shown in the supplementary material.

To create a population level exposure series for particulate matter, we used the centering approach described by Schwartz (2000). For each monitor, the mean (overall observations) of that particular monitor was subtracted from each observation. This demeaned data was then divided by the standard deviation of that particular monitor to get a standardized series for that monitor. This process was repeated for all monitors in a given city. The standardized series across all monitors was averaged to get one single series. Finally, this single series was multiplied by the standard deviation of all monitors taken together and the mean of all monitors taken together was added back to each observation (Schwartz, 2000). The resultant series was the final exposure series used in the regression model.

After creating the final exposure series, we dropped all observations where pollution data was not available (i.e. complete case analysis). For this dataset, the final exposure series for PM_{10} was shifted (lagged) by one observation. This would relate the deaths on a given day to a PM value roughly three days prior (i.e. for instance deaths on June 4 are associated with pollution value measured on June 1; deaths on June 7 are associated with pollution value measured on June 4 and so on).

2.3. Analytical models

We adopted a semi-parametric regression framework to develop the exposure – response relationship between air pollution and mortality for the sampled cities (Balakrishnan et al., 2011; Peng et al., 2006; Rajarathnam et al., 2011). The logarithm of daily expected deaths was modelled as a function of daily air pollution measurements in the presence of other confounding variables such as temperature and humidity. We assumed deaths to follow an over-dispersed Poisson distribution i.e. $E(Y_t) = \mu_t$ and $\mathrm{Var}(Y_t) = \phi\mu_t$ where ϕ is the over–dispersion parameter. This is an accepted assumption for pollution studies (Dominici et al., 2004). Smooth functions were used to control for effects of daily temperature, humidity and seasonal and long term trends as these are non-linearly related to mortality (see supplementary material figures). Thus the regression equation can be expressed as:

$$\mathrm{Log}\left[E\left(Y_{ij}\right)\right] = \beta \mathrm{PM}_{10i,j-1} + \sum_{j=1}^{P} g(x_{ij}) + \mathrm{DOW}_{ij} + H_{ij} + \varepsilon_{ij} \tag{1}$$

where Y_{ij} is the daily mortality count for the ith city on the jth day and is assumed to follow an over-dispersed Poisson distribution. The pollution (PM_{10}) measurement for the ith city on the jth day lagged by one observation is represented using $\beta PM_{10i,j-1}$. The covariates x_{ij} represent daily temperature, relative humidity and time for the ith city on the jth day. The effects are expressed by an unknown smooth function $g(\bullet)$ constructed using natural cubic splines. Details about the structure of $g(\bullet)$ are given in the supplementary material. An indicator variable for each day of week is given by DOW_{ij}. Public holidays were controlled for using an indicator variable given by H_{ij}. The error term is modelled using ε_{ij}. The parameter of interest is β associated with the pollution variable (PM_{10}). This parameter gives an estimate of the increase in mortality associated with a unit change in PM_{10} concentrations.

For temperature, humidity and time, the amount of smoothness (i.e. optimal degrees of freedom) was determined based on the approach by Dominici et al. (2004). The underlying idea is that β is sensitive to degrees of freedom selected for temperature, humidity and time. The approach by Dominici et al. (2004), where optimal degrees of freedom are chosen such that they predict PM_{10} instead of daily mortality, provides asymptotically unbiased estimates of the β parameter. The details of the algorithm implemented to arrive at these optimal values have been provided as supplementary material.

Mortality impacts related to pollution may be delayed i.e. exposures on preceding days may determine current health outcomes (Bhaskaran et al., 2013; Braga et al., 2001). The lack of daily PM_{10} measurements did not allow for use of distributed lag models as this may introduce large errors (Braga et al., 2001; Zanobetti et al., 2000). Instead we lagged the exposure series by one observation as suggested by Balakrishnan et al. (2011).

2.4. Sensitivity analysis

In order to compare some plausible scenarios, a sensitivity analysis was undertaken where the estimates (β) were tested using (i) zero lags for the pollution variable; (ii) minimum temperature instead of maximum temperature and (iii) including other pollutants such as sulphur dioxide.

2.5. Temperature – pollution interactions

To study the interaction effects of temperature and pollution (PM_{10}) on mortality, we used two approaches. The first approach, suggested by Ren et al. (2006), involved fitting Equation (1) with an interaction term to capture the joint effects of pollution and temperature. This model is given in Equation (2). The term $t_{ij}{*}PM_{10ij}$ expresses the interaction between daily temperature and pollution while its effect is quantified by the coefficient α.

$$\mathrm{Log}\left[E\left(Y_{ij}\right)\right] = \beta \mathrm{PM}_{10i,j-1} + \sum_{j=1}^{P} g(x_{ij}) + \alpha\left(t_{ij}{*}\mathrm{PM}_{10ij}\right) + \mathrm{DOW}_{ij} + H_{ij} + \varepsilon_{ij} \tag{2}$$

If the interaction term (α) is found to be significant, then a second model is used to understand if interaction effects are more significant during hotter or colder temperatures.

The second approach, described by Chen et al. (2014), involves dividing temperature into different levels and then studying the temperature – pollution interaction for each level. For each city, we divided the temperature into four quartiles. We then estimated Equation (2) for each of the four quartiles.

2.6. Software

All analysis was performed in the statistical environment R version 2.15.1. The package mgcv (version 1.7–24) was used to fit the models described in Equations (1) and (2). The package *ggplot2* (version 0.9.3.1) was used for graphical representations.

3. Results

3.1. Summary statistics

As seen in Table 2, there is wide variation among different cities when it comes to daily pollution levels, mortality, temperature, as well as number of complete observations available for analysis. The highest PM_{10} levels are observed for Mumbai (174.4 ± 86.6) and the lowest for Shimla (54.4 ± 25.2). The daily number of deaths varies across cities and is linked to population size. Shimla had the lowest number of daily deaths (4.2 ± 2.7) whereas Mumbai (225.6 ± 30.7) had the highest.

Each city had a different number of air quality monitors. The number of air quality monitors ranged from one in Shimla to nine in Hyderabad. For every air quality monitor, percentage of missing data by year varied (see Table S1 in supplementary material). Air pollution impacts were estimated for the period of 2008–09 for Hyderabad and Bangalore; from 2005 to 2009 for Ahmedabad; from 2005 to 2011 for Mumbai and 2006–2009 for Shimla.

3.2. Exposure – response estimates

The percentage increase in mortality associated with a 10 $\mu g/m^3$ increase in PM_{10} is reported in Table 3. The highest increase was seen for Shimla (1.36%) and the least for Ahmedabad (0.16%). Bangalore and Mumbai showed similar results with a 0.22% and 0.20% mortality increase respectively. The dosage–response curves for each city are provided in the supplementary material (Figure eS.13 to eS.17).

The sensitivity analysis showed that mortality estimates were lower when no lag for pollution was used, across all cities. The estimates of the core model did not change significantly if minimum temperature was used as a confounding variable. The inclusion of SO_2 reduced the impact of PM_{10} on mortality for Hyderabad and Mumbai, although, these differences were not significant. In addition, no significant interaction effect (at a 5% level) between temperature and pollution on mortality was observed. Table 4 and Table 5 show the estimates, standard errors and p-values for the interaction term between temperature and pollution.

3.3. Comparison with other studies

Our results are in close agreement with previous studies (Balakrishnan et al., 2011; Rajarathnam et al., 2011; Romieu et al., 2012), which find 0.44% (95% CI = 0.17–0.71) for 0.15% (95% CI = 0.07–0.23) increase in mortality for every 10 $\mu g/m^3$ PM_{10} increase for Chennai (Balakrishnan et al., 2011), and Delhi (Rajarathnam et al., 2011), respectively. The APHENA Study examined associations between PM_{10} and mortality as well as hospital admissions across multiple cities in United States, Europe and

Table 2
Observed values for different variables across cities (mean ± standard deviation).

City	Temperature (°C)		Relative humidity (%)	Daily deaths	PM_{10} (μg/m^3)	No of complete observations
	Maximum	Minimum				
Ahmedabad	34.3 ± 4.6	21.5 ± 5.6	56.7 ± 17.8	100 ± 18	93.9 ± 58.7	602
Bangalore	29.4 ± 2.7	18.7 ± 2.3	66.6 ± 15.5	120.7 ± 17	108.3 ± 69.8	307
Hyderabad	33.6 ± 3.8	20.7 ± 3.6	53.1 ± 17.6	74.7 ± 16.2	80.4 ± 21.9	498
Mumbai	32.3 ± 2.4	22.7 ± 4.05	69.2 ± 13.8	225.6 ± 30.7	174.4 ± 86.6	2012
Shimla	20 ± 5.1	11.2 ± 5.3	NA	4.2 ± 2.7	54.4 ± 25.2	962

*NA – No data available.

Table 3
Percentage increase in mortality for every 10 μg/m^3 PM_{10} increase.

	Core model	Sensitivity analysis		
		No lag for PM	Minimum temperature	SO_2 included
Ahmedabad *(95% CI)*	0.16% *(−0.31 to 0.62)*	0.06% *(−0.42 to 0.55)*	0.12% *(−0.36 to 0.61)*	−0.32% *(−1.01 to 0.38)*
Bangalore *(95% CI)*	0.22% *(−0.04 to 0.49)*	0.16% *(−0.11 to 0.43)*	0.17% *(−0.09 to 0.43)*	0.23% *(−0.18 to 0.64)*
Hyderabad *(95% CI)*	0.85% *(0.06 to 1.63)*	0.48% *(−0.30 to 1.27)*	0.83% *(0.04 to 1.62)*	0.41% *(−0.69 to 1.51)*
Mumbai *(95% CI)*	0.20% *(0.10 to 0.30)*	0.18% *(0.08 to 0.27)*	0.16% *(0.06 to 0.25)*	0.13% *(0.04 to 0.23)*
Shimla[a] *(95% CI)*	1.36% *(−0.38 to 3.1)*	0.97% *(−0.83 to 2.7)*	0.54% *(−1.17 to 2.27)*	–

Negative values imply that the effect of pollution on mortality is not significant.
[a] No humidity and SO_2 measurements were available for Shimla; Values in the brackets represent 95% confidence intervals.

Table 4
Interaction effects of temperature and pollution for all cities (based on approach by Ren et al., 2006).

City	β co-efficient	Std. error	p – value
Ahmedabad	0.00328	0.00202	0.11
Bangalore	0.00607	0.00473	0.20
Hyderabad	0.00759	0.00757	0.31
Mumbai	−0.00380	0.00281	0.16
Shimla	−0.00007	0.00054	0.89

Table 5
Interaction effects *(p-value)* and [range] across different temperature levels for all cities (based on approach by Chen et al., 2014).

	Ahmedabad	Bangalore	Hyderabad	Mumbai	Shimla
Lowest Quartile	−0.00005 *(0.81)* [20.3 −31 °C]	0.00077 *(0.05)* [21.2 −27.5 °C]	0.00077 *(0.28)* [22.6 −30.7 °C]	−0.00012 *(<0.01)* [19.8 −30.8 °C]	−0.00027 *(0.68)* [0.6–17 °C]
Second Quartile	−0.00053 *(0.37)* [31.1 −34 °C]	−0.00105 *(0.08)* [27.6 −29.3 °C]	−0.00060 *(0.42)* [30.8 −33.1 °C]	0.00013 *(0.39)* [30.9 −32.5 °C]	−0.00215 *(0.45)* [17.1 −21 °C]
Third Quartile	0.00009 *(0.85)* [34.1 −37.2 °C]	0.00007 *(0.92)* [29.4 −30.7 °C]	0.00158 *(0.25)* [33.2 −35.9 °C]	−0.00009 *(0.39)* [32.6–34 °C]	−0.00106 *(0.43)* [21.1 −23.4 °C]
Highest Quartile	−0.00033 *(0.29)* [37.3 −44.4 °C]	0.00013 *(0.48)* [30.8 −36.7 °C]	−0.00017 *(0.83)* [36 −43.4 °C]	0.00004 *(0.53)* [34.1 −41.3 °C]	−0.00089 *(0.32)* [23.5 −31 °C]

Canada. They found that average all cause mortality risks associated with PM_{10} across all ages were 0.36% (95% CI = 0.21–0.50) for Europe, 0.38% (95% CI = 0.26–0.50) for United States and 1.1% (95% CI = 0.58–1.6) for Canada (Samoli et al., 2008). These findings imply that the short term mortality risks associated with pollution are not very different across cities. However, exposure of the large Indian population to outdoor pollution translates into a significant increase in mortality. Fig. 1 compares the percentage change in mortality for every 10 μg/m^3 increase in PM_{10} across different studies.

4. Discussion

Our study was designed to explore the differences in short term impacts of air pollution on mortality and possible modification due to temperature in Indian cities that are spread across climactic zones and topographies. The results add to the existing body of epidemiological knowledge in the context of a developing country, like India.

An interesting finding of our analysis is that cities such as Ahmedabad and Mumbai that have higher levels of pollution experience a relatively lower increase in mortality for every 10 μg/m^3 increase in PM_{10}. In contrast, the percentage increase in mortality is highest for Shimla which is among the cleanest cities. These results are corroborated by recent studies that have attempted to develop a new set of exposure – response relationships linking pollution and health (Lim et al., 2013; Smith et al., 2013). One of their main findings was that the impact of pollution on health may indeed be non-linear in nature i.e. when baseline pollution levels are high, health benefits associated with reduction in PM_{10} may be very small as compared to a situation where baseline pollution levels are low (Lim et al., 2013; Smith and Peel, 2010; Smith et al., 2013).

The import of these findings is that small reductions in pollution in cleaner cities will yield large health benefits, whereas in less cleaner cities, even large reduction in pollution may yield only modest health benefits in a relative sense. However, it is not to suggest that the focus should be on reducing pollution in cleaner cities alone. On the contrary it underscores the need for rapid and aggressive policy measures in both types of cities to curb air pollution. Ambitious targets towards achieving ambient air quality standards should be set in highly polluted cities. On the other hand, cleaner cities could leverage significant health gains even by focussing on small reductions in pollution.

The pollutant of choice in our study was particulate matter less than 10 μg in size (PM_{10}). This is because it is the most routinely monitored air pollutant in India. However, there remains the need to expand routine monitoring to other pollutants such as $PM_{2.5}$, black carbon, ozone, benzene, carbon monoxide, polycyclic aromatic hydrocarbons and heavy metals.

The differential health impacts of single versus multiple pollutant models are of interest in epidemiology, although it is unclear whether including more than one pollutant in the analysis is necessarily more beneficial as opposed to single pollutant models (Tolbert et al., 2007). We focussed primarily on the impacts of PM_{10} on mortality. Inclusion of sulphur dioxide (SO_2) along with PM_{10} did not change our estimates significantly, similar to previous

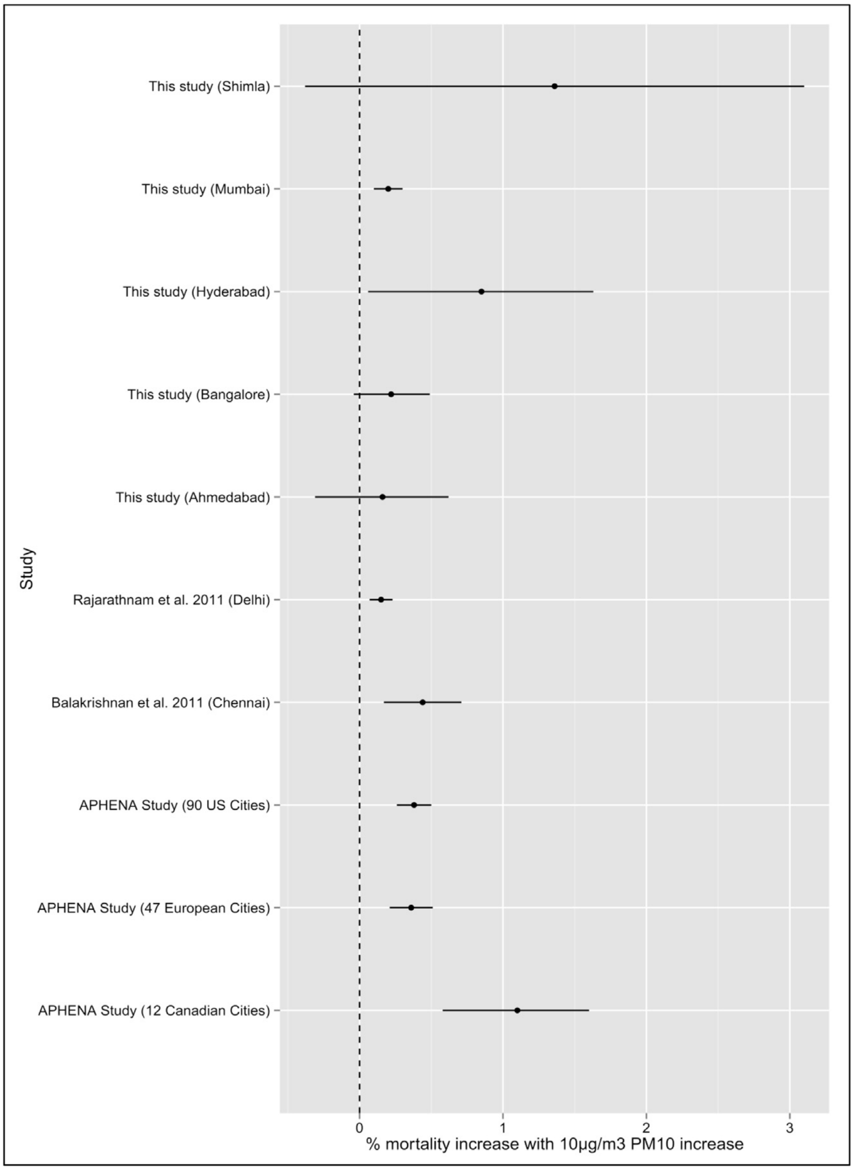

Fig. 1. Shows the central estimate and 95% confidence intervals for percentage increase in all cause mortality with every 10 μg/m^3 increase in PM_{10} at Lag 1. We compare estimates for five cities analyzed in this study with those from selected previous studies.

findings (Rajarathnam et al., 2011). High percentage of missing data precluded incorporating nitrous oxides (NO_x) in our modelling framework. This is a limitation of our study and remains an important area of future studies and research.

In the context of our modelling framework, we did not find significant impacts of temperature-pollution interactions on mortality for the cities studied. This may be because temperature – pollution interactions are highly complex and non-linear and therefore may not have been captured adequately in the current model framework. Some studies e.g. Ren et al. (2006), have used more complex approaches such as modelling of the interaction using locally weighted smoothing functions (LOESS). However, a key limitation of such complex models is the inability to interpret estimates in an intuitive manner. Furthermore, the results from previous studies are varied implying that interaction effects may be city specific in nature. For instance, within the United States alone, Ren et al. (2008), found that while ozone modified the temperature mortality relationship in northern cities, no such effects were observed for southern cities. Further research is needed to better understand how temperature and pollution interactions influence health risks across cities in India.

One key limitation of our dataset is that there were significant missing data for the different air quality monitors across cities except Mumbai. This affected the parameterization and structure of the semi-parametric model used in our analysis. Furthermore, the fact that pollution estimates were not significant for Ahmedabad and Shimla may be a reflection of measurement error. Needless to say, better monitoring will help in developing more accurate exposure – response relationships across cities.

To create a consistent exposure series, Balakrishnan et al. (2011), developed a spatial model for Chennai. A 0.5 square kilometre grid was superimposed on a map of zones in the city. For each grid cell, PM values of the nearest AQM (measured as distance from centroid of grid to AQM) were assigned. For each zone, the PM exposure series was an average of the air quality reading on a particular day weighted by the number of grid cells it was assigned to in the zone.

This approach was preferred to a simple average or centering approach used in this study. Whereas, a spatial model has distinct advantages, it requires a large amount of disaggregated information such as daily number of deaths in different zones of the city. Since that information was not available for cities in the current study, a centering approach was adopted. The advantage of the centering approach is that although a difference in measurements across monitors may influence variability of the exposure series and lead to underestimates, the slope co-efficient (i.e. β) corresponding to the pollution parameter (i.e. PM_{10}) remains unchanged if one or several AQM's are used (Balakrishnan et al., 2011; Rajarathnam et al., 2011; Wong et al., 2001).

Though it has been pointed out that impacts of air pollution is primarily linked to cardio-respiratory mortality (Pope III et al., 2002; Samoli et al., 2014; USEPA, 2009), the present study only examined all-cause mortality. This was because information on cause-of-death and age groups was not available for the cities which were considered. Mortality in India is underreported and on an average only 67% of all deaths gets registered, with high variability across different states (Dhar, 2013). Of these, it is only institutional deaths that contain information on cause of death. This remains a limitation of the study. It is vital for future studies to enhance the quality of mortality registration data in India.

In conclusion, the study of air pollution on mortality remains an important area of research in the Indian context. Clearly there remains a need to strengthen data quality and carry out similar studies for many more cities. In addition to the time-series approach used in this study, cohort studies are required to understand air pollution related health risks in India. Epidemiological evidence can help guide policy by providing evidence to tightly couple health outcomes and air quality standards, thereby minimizing the impacts of outdoor air pollution in India.

Author contributions

H.H.D and A.G. were responsible for research design, methodology and interpretation of results. H.H.D and D.B. carried out statistical analysis. All authors contributed to writing the manuscript.

Financial interests' declaration

None declared.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.09.071.

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