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Differential toxicity of PM2.5 components and modified health effects modeling: A case study in Nepal

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Differential toxicity of $PM_{2.5}$ components and modified health effects modeling: A case study in Nepal

A Thesis Presented
by
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ABSTRACT

DIFFERENTIAL TOXICITY OF $PM_{2.5}$ COMPONENTS AND MODIFIED HEALTH EFFECTS MODELING: A CASE STUDY IN NEPAL

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During the latter part of the 20th century, a transition away from coal as a major energy source in developed countries was accompanied by a notable decrease in air pollution-related deaths in those countries. Currently the same phenomenon is being observed in developing nations like China and India. However, many areas that do still rely on coal for their energy production or industrial needs also reflect a gap in research on the effects of those specific processes on local populations. Located in Nepal at the foot of the Himalayan Plateau, Kathmandu represents one such location. The local economy of Kathmandu and the surrounding area relies heavily on the production of bricks using coal-fired kilns, which produce large amounts of particulate matter. This particulate matter contains a characteristic mix of metals. This unique fingerprint can be used to identify and track kiln emissions in ambient samples. We collected hourly samples of ambient metal concentrations over a period of three months at the start of 2019. We then used these data to perform positive matrix factorization (PMF) to identify several factors contributing to the ambient air pollution of the sampled area, each representing a source type. The PMF output included the chemical 'fingerprint' of each factor as well as hourly variation of each factor. We were able to isolate the fraction of $PM_{2.5}$ contributed by coal and estimate the health effects attributable to this fraction using a modified risk ratio of 1.05 to

reflect the higher toxicity of coal emissions. We found that the current estimates of health impacts in Nepal underestimate the true impact of coal by 416 deaths per year.

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CHAPTER 1

INTRODUCTION

Nepal is a landlocked country in South Asia with a population of approximately 30 million people and a total area of 143,350km^2 (WorldBank, 2018). Bordered by China to the north and by India to the south, east, and west, Nepal's terrain is a mixture of mountains and low-lying valleys; its elevation ranges from less than 100 meters above sea level to the highest point on earth at 8,848 meters above sea level. Nepal has a GDP of 29.04 billion USD of which agriculture accounts for nearly one third, and industry another 15% (WorldBank, 2018). Brick manufacturing represents a fraction of industry that is not yet represented as a formal sector, despite bringing in 37 million USD in investments.

The brick industry has been faced with social and environmental issues in recent years due to the heavily polluting nature of the brick manufacturing process, for which coal is the main fuel stock. Burning coal is a process that produces fine airborne particles and coal dust, as well as the gaseous precursors to smog, acid rain, and ground-level ozone. Brick kilns produce huge amounts of air pollution, consuming about 450,000 tons of coal and emitting 1.1 million tons of CO2 each year (Maskey et al., 2013). The industry is especially hazardous for brick kiln workers, whose additional exposures include carbon monoxide and airborne silica dust (Thygerson et al., 2016). In 1997, Worldbank found brick kilns to be the second highest source of particulate matter in Nepal.

Airborne particulate matter (PM) is a broad class of pollutants that have been of growing concern to environmental health researchers since the mid-20th century. PM is usually subdivided into specific designations based on particle diameter, the most common designations being PM_{10} and $PM_{2.5}$. These designations are so named based on the aerodynamic particle diameter; $PM_{2.5}$ refers to all suspended particles in a given sample with an aerodynamic diameter of 2.5µm or less. Notably, this means that all particles measured as $PM_{2.5}$ will also be counted in a measurement of PM_{10}. The larger particles measured in PM_{10} tend to be generated through mechanical weathering and erosion, such as fine dust blown around by wind. The two main mechanisms through which $PM_{2.5}$ is generated is by direct emission of particles from combustion, and from secondary formation from gaseous precursors.

Health concerns associated with PM_{10} tend to be respiratory in nature, related to the deposition of particles in the airway and lungs. Many of the larger particles can be filtered or removed by the body's natural processes.

$PM_{2.5}$ is associated with more serious health risks involving the cardiovascular system in addition to respiratory effects. This is because $PM_{2.5}$ is small enough to penetrate deeply into the lungs, where it can enter the bloodstream and cause damage to the heart, lungs, blood vessels, and other organs. Some known related health outcomes include heart disease, lung cancer, and hypertension. Because it is not typically acutely toxic, it can be difficult to attribute real-life health outcomes to exposure to $PM_{2.5}$, and epidemiological analysis is required.

In the United States, the Clean Air Act (1970) set the groundwork to establish standards for 6 criteria air pollutants to be monitored by the Environmental Protection Agency in order to maintain clean and healthy air quality for Americans. One of these pollutants is particulate matter, the standards for which are subdivided into individual standards for PM_{10} and $PM_{2.5}$. The primary standard for $PM_{2.5}$ requires that the annual mean concentration of ambient $PM_{2.5}$ be less than 12µg/m^3 averaged over 3 years. Locations that exceed this standard are considered in non-attainment. States also have the ability to set their own standards and regulatory bodies such as the California Air Resources Board (CARB), which sets standards for California that are also followed by several other states, though these standards are required to be at least as stringent as comparable federal standards.

1.1 Coal in Nepal

There are currently an estimated 1000 coal-fired brick kilns in operation in Nepal (*Nepal Energy Sector Assessment, Strategy, and Roadmap*, 2017). The exact number is uncertain because although there is a registry that exists, unregistered brick kilns are common and pose a threat to the development of the industry due to their lack of regulation. Nepal is mostly dependent on India for its coal, with only about 5% produced domestically. Local lignite coal from the Dang region of Nepal's Lumbini province is typically not as energy efficient as imported coal, and local coal producers will mix local coal with coal imported from Assam and Bihar provinces in India in a 50-50 mixture. Brick producers are often unaware of the quality of the coal they are using to fuel their kilns (*Brick Sector in Nepal National Policy Framework*, 2017). In Kathmandu Valley about 100 brick kilns are responsible for 40% of the PM_{10} present, making up the largest contribution of any single identifiable source (B. M. Kim et al., 2015; Maskey et al., 2013).

Nepalese brick kilns are moving-fire continuous kilns that rely on the flow of air to carry fire through green (unbaked) bricks stacked between the outer and inner walls of the kiln. These are natural draught kilns in which the chimney provides the draught necessary to maintain the correct air flow and temperature in the kiln. Green bricks are stacked in the firing zone closest to the center of the kiln where temperatures are hottest, and baked bricks are stacked near the outer wall of the kiln where cool air entering the chamber maintains the correct temperature for cooling. Once ignited, kilns burn continuously for a matter of weeks while bricks inside are shifted to maintain the correct temperature for different stages of baking.

Bull's Trench Kilns are the most common coal firing technology, with at least 950 kilns of this design operating in the country as of 2017. Within this designation, kilns can be categorized into one of two categories based on their brick-setting strategies. Straight line kilns allow for air movement in a straight line through the kiln, whereas zigzag kilns force the air to move in a zigzag pattern. Zigzag kilns demonstrate more efficient heat transfer to the bricks and increased scavenging of particles on kiln walls, making them generally cleaner and more efficient (*Brick Sector in Nepal National Policy Framework*, 2017).

a) ***b)***

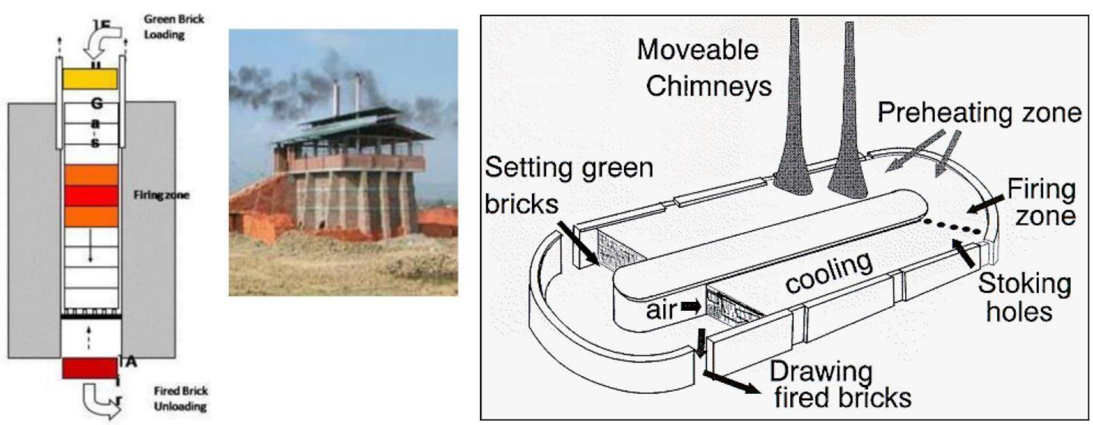

Figure 7: kiln diagrams taken from (Thygerson et al., 2016)
a) Vertical Shaft Trench Kiln (VSTK) Layout. Green bricks are loaded into the top and slowly move down into the firing zone. Unloading occurs at the base. Less heat loss and better heat exchange makes this technology most efficient
b) Movable Chimney Bull Trench Kiln (MCBTK) Layout. The circular trench holds roughly 200 - 300,000 bricks at a time. The chimneys made of sheet metal are moved around as the firing progresses and stabilized using wiring attached to the top

Other kiln technologies include Vertical Shaft Brick Kilns, Tunnel Brick Kilns, Hoffman, and Hybrid-Hoffman kilns, which together represent only 35 kilns currently in operation (*Brick Sector in Nepal National Policy Framework*, 2017). While Vertical Shaft Brick Kilns are the most efficient and environmentally friendly technology available, emitting ten to twenty times less black carbon than Bull's Trench Kilns, widespread adoption of this technology has been slow due to the inferior quality of bricks (Premchander et al., 2011). In addition to black carbon, kilns emit sulfur dioxide (SO_2) and carbon dioxide (CO_2), as well as small amounts of As and Ca.

1.2 Environmental Policy in Nepal

			Concentration in Ambient Air, Maximum		
Parameters	Units	Average Timing	Nepali National Standards	EPA Standards	WHO AQG
TSP	µg/m³	24-hrs	230	-	-
PM_{10}	µg/m³	24-hrs	120	150	45
Sulfur Dioxide	µg/m³	annual	50	-	-
		24-hrs	70	196.5*	40
Nitrogen Dioxide	µg/m³	annual	40	100	10
		24-hrs	80	-	25
$PM_{2.5}$	µg/m³	24-hrs	40	35	15
Lead	µg/m³	annual	0.5	0.15*	-
Carbon Monoxide	µg/m³	8-hrs	10,000	10,305	-
Ozone	µg/m³	8-hrs	157	-	100
benzene	µg/m³	annual	20	-	-

Table 2: Nepali National Air Quality Standards, EPA National Ambient Air Quality Standards, and WHO Air Quality Guidelines.
**US NAAQS require a 1-hour averaging time for this primary standard.*
***US NAAQS require a 3-month rolling average for Lead Concentrations.*

Nepal currently has national Ambient Air Quality Standards (NAAQS) that were instated in 2012 and are enforced by the Ministry of the Environment under authority granted by rule 15 of the Environmental Protection Regulations. These standards, shown in table 1 alongside WHO guidelines, set a 24-hour standard for $PM_{2.5}$ of 40 µg/m^3, requiring that the average concentration of a set of samples taken in a location over 24 hours remain under this level to be considered in attainment. In addition to NAAQS and under the same authority, the Ministry of the Environment had previously published the Standard on Chimney Height and Emission for Brick Kilns in 2008. These standards set individual minimum chimney height in meters from the ground for kilns and maximum total particle concentration measured at the point of emission for three kiln types. It is unclear how these standards are enforced, or how kilns outside of those three categories are regulated.

1.3 $PM_{2.5}$ Health Impacts

Exposure to airborne particulate matter (PM), and especially $PM_{2.5}$, is ranked as the fifth-highest mortality risk factor globally. In 2015, $PM_{2.5}$ exposure accounted for an estimated 7.9% of total deaths globally and 4.2% of total disability-adjusted life-years; 59% of these being in east and south Asia (Cohen et al., 2017). $PM_{2.5}$ is associated with cardiovascular disease (CVD) and respiratory issues including certain kinds of cancers (Amsalu et al., 2019; Guaita et al., 2011; Guo et al., 2020). CVDs are the leading cause of death globally, with ischemic heart disease (IHD) accounting for more than half of all CVD deaths in 2016 (*Ambient Air Pollution: A Global Assessment of Exposure and Burden of Disease*, 2016). The World Health Organization (WHO) reported annual median concentration of $PM_{2.5}$ to be as high as 140µg/m^3 in urban areas of Nepal; this is 10 times higher than the recommended level (*Ambient Air Pollution: A Global Assessment of Exposure and Burden of Disease*, 2016). Furthermore, in a study looking at the exposure of brick kiln workers to $PM_{2.5}$, concentrations of PM inside workers homes were found to be not significantly different from outdoor concentrations, meaning that this exposure is nearly constant (Thygerson et al., 2019).

Particulate air pollution in Kathmandu is a complex mixture of primary and secondary emissions from a variety of sources, which themselves vary in the risk posed to human health (B. M. Kim et al., 2015; Thurston et al., 2016). To fully and accurately understand the level of risk associated with ambient particulate air pollution, we must first understand the properties of each major source type contributing to this mixture, and the role they each play in human health.

1.4 Source Apportionment

Source Apportionment is the application of mathematical models to analyze the co-variability of pollution components and assign them to factors representing real-life emission sources; it is a crucial first step to developing measures to control and reduce the impact of pollution on a population. Understanding which sources drive contamination can help researchers understand not only the best methods of reducing emissions, but also give a better understanding of the effects that pollution is having on the population. Source apportionment has been widely used to study air pollution over the past few decades, but it has also been applied to soil and groundwater pollution (Diakite et al., n.d.; B. M. Kim et al., 2015; E. Kim et al., 2005).

After a comprehensive review of the current literature regarding air pollution in the Kathmandu Valley, we found previous source apportionment studies have been conducted using 24-hour sampling techniques (B. M. Kim et al., 2015; E. Kim et al., 2005). However recent studies have shown that higher time-resolution in sampling allows for robust source apportionment due to the greater variation captured in the analysis (Q. Wang et al., 2018). This represents a knowledge gap in the literature that this study aims to fill. With this information, we will be able to create a more accurate picture of airborne $PM_{2.5}$ in Kathmandu Valley and its health effects on the population and allow lawmakers to make more informed legislative choices regarding the health of the Nepalese people.

Our first main aim is to conduct source apportionment and to identify and quantify each of the resulting source types (coal burning, crustal material, industrial emissions, etc.). Once a coal signal has been identified, we aim to identify how frequently brick kilns emissions drive aerosol in Kathmandu by comparing the intensity of the coal signal to ambient PM2.5 concentrations. Our second main aim is to compare this empirical result to assumed mortality risk in AirQ+ by modifying IER ratios to reflect coal emissions. According to Thurston et al., coal PM has a hazard ratio for IHD equal to 1.05 (95%CI: 1.02, 1.08) when compared to ambient PM (Ito et al., 2013; Thurston et al., 2016). Finally, we aim to quantify lives saved if brick kiln emissions ceased or were dramatically altered.

CHAPTER 2

METHODS

Data collection took place between 01-08-2019 and 3-15-2019 in Bode, located in the Kathmandu Valley about 8 miles from the city of Kathmandu, Nepal, using an XACT620 (Cooper Environmental). This instrument collects and analyzes hourly samples of PM2.5 to calculate the average concentration of specific elements present in the sample over the last hour. Our data included 1036 hourly samples of ambient aerosol, which were further analyzed for up to 23 different elements. Data processing was computed in R Studio, using packages Openair, Lubridate, and dplyr.

2.1 Sample Collection

Hourly chemical speciation data was collected using an XACT620 from Cooper Environmental Services (Beaverton, USA). This instrument pumps ambient air at a constant rate first through a cyclone to inertially remove particles greater than 2.5 µm in diameter, and particles are then deposited on a reel-to-reel polyflourotetraethylene (PFTE) filter tape. After 1 hour of sample collection, the filter tape is automatically advanced, and the sample of particulate matter is irradiated by an x-ray beam. The fluorescence of the sample is measured by a self-contained X-ray fluoroscope, and the instrument calculates the mass of each calibrated element present in the sample. The onboard computer divides the mass by the volume of air pumped during the hour of sampling to calculate the average ambient concentration of each element present in PM2.5 during that hour. This instrument provides hourly time series data of air pollution constituents, with which we were able to conduct our source apportionment analysis using Positive Matrix Factorization (PMF)

2.2 Positive Matrix Factorization

PMF is a tool designed by the US EPA to estimate the contributions to an environmental sample made by specific sources using the chemical composition of the source. The program uses receptor models to solve the chemical mass balance (CMB) represented by equation 1-1, applied to a dataset x of i by j dimensions, with i number of samples and j number of species, measured with uncertainties u, with p number of factors, species profile f of each source, and g mass contributed by each factor to each individual sample:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij}$$

e_{ij} represents the residuals of each sample and species. PMF works to solve the CMB by minimizing Q in objective equation 1-2:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right]^2$$

PMF begins its search for optimal factors and contributions by choosing a random factor and systematically modifying this factor to continually reduce Q and approach the best-fit solution. Because of the random nature of the starting point, the model will sometimes end on a local minimum as opposed to the global minimum, and as such should be run multiple times to maximize the chance of landing on the global minimum.

From the input chemical speciation matrix, PMF constructs a matrix of factor contributions (F) and a matrix of factor profiles (G). The factor contribution matrix displays the relative contribution of factors to each sample, normalized across all samples for each factor.

This allows the user to create a time-series and observe the changes in factor expression over time. The factor profile matrix contains the average mass concentration of each chemical species contributed by each factor, which can be interpreted by the user to determine the real-life source category represented by each factor. With these two datasets, the user can track the relative changes in expression of identifiable source categories across the sampling period.

2.3 AirQ+

AirQ+ is software developed by the World Health Organization to model the impacts of air pollution on specific populations. The software computes excess mortality risk attributed to air pollution exposures across several health endpoints, and the analysis is based on GBD literature values of risk across age categories. We use this tool to calculate excess risk within a specific population and integrate these values to estimate excess mortality in Nepal.

To complete a burden of disease analysis, AirQ+ requires air quality data, population data, and data regarding the health outcome of interest. Air quality data can be entered as a daily time series or a single mean concentration and must be entered as $\mu g/m^3$. Population data requires both the total population and the at-risk population for the specific health endpoint.

To make burden of disease calculations, AirQ+ uses standard Integrated Exposure Response (IER) functions, which are based on the Global Burden of Disease study 2015-2016. For example, risk Values for Ischemic Heart Disease (IHD) provided in AirQ+ represent cumulative lifetime exposure risk that consider duration of exposure; air pollution is more likely to eventually cause a chronic health outcome in a young person because that person may experience 50+ years of exposure in their lifetime. Risk values are also non-specific for PM2.5 and ignore the origin and chemical composition of the pollutant. According to Thurston et al., coal PM has a hazard ratio for IHD equal to 1.05 (95%CI: 1.02, 1.08) when compared to ambient

PM. Because we are investigating mortality in Nepal, where aerosol composition can be quite different, we modified the original IER tables with these values to reflect the additional risk posed by coal, and used these modified tables in AirQ+ to calculate the effects of coal PM.

We sourced our CVD mortality data from Bhattarai et al. and population data from Worldbank (Bhattarai et al., 2020). AirQ+ allows for burden of disease calculations to be performed in 5-year age brackets starting at age 25 up to a 95+ age bracket including all individuals 95 and over. CVD mortality data is available in corresponding 5-year age brackets, apart from a single age group encompassing all individuals 80 and older. Because mortality data for this 80+ age group is not further broken down into 5-year brackets, burden of disease calculations for this group had to be made using an IER that was not representative of the entire group. We chose to perform these calculations using the 80-84 IER, as the plurality of 80+ individuals likely fall in this range. This will cause an overestimation of attributable health effects in this age bracket but is unlikely to impact the validity of our results because it only represents 0.74% of the total population.

In calculating the health effects of coal in the Kathmandu Valley, we have two goals, the first of which is to calculate the total number of deaths attributable to coal PM. This will represent the number of deaths that would be prevented if coal-burning kilns were replaced with non-emitting technology. The second goal is to calculate the total number of deaths attributable to the increased toxicity of coal PM controlling for total PM concentration. This value would represent the total number of annual deaths that would be prevented if coal-burning kilns were replaced with similarly emitting, non-coal kilns.

Our first goal was achieved by running AirQ+ using the daily coal PM concentrations calculated in PMF to directly estimate the number of deaths attributable to coal PM. These concentrations were calculated on 47 days ranging from 1.22 $\mu g/m^3$ to 14.66 $\mu g/m^3$, with a mean of 6.87$\mu g/m^3$. It should be reiterated that these bulk concentrations only reflect a fraction of ambient PM in the region, as the data are limited to only the metal concentrations sampled by our instrumentation. PM is comprised of a wide range of chemical species, many of which cannot be observed by XRF detection, most notably carbonaceous materials and ions, and as such we expect the total concentration of PM to be much higher, and for PM attributed to coal to be only a fraction of total PM. As such, the average concentration of a factor calculated by PMF is of little use by itself for the purposes of this study, but rather the contribution of that factor to the total PM calculated across all elements sampled. That contribution, when scaled to the total $PM_{2.5}$ concentration measured at the US Embassy, provides the most accurate estimate of factor expression available to us.

To calculate the effects of the increased toxicity of coal PM, we subtracted two estimations of PM-attributed annual mortality, the first representing our current estimations of PM2.5 health effects that do not consider the toxicity of coal PM, and the second representing a "worst case" scenario in which PM is 100% coal and remains at peak level year-round. We then adjusted the remainder using a model we constructed of yearly PM variation and coal contribution. The final product of these calculations being the estimated number of annual deaths attributable to the increased toxicity of coal.

CHAPTER 3

RESULTS

Our sampling in Nepal yielded time series data about the concentration of each of the 24 elements sampled. This time series data was used to construct four factors in PMF, which we were able to identify and confirm using several different lines of reasoning. Once a coal factor had been identified, we used this data to make estimations about the true health impacts of coal emissions in Nepal.

3.1 Assigning Sources in PMF: Identifying Factors

As in any urban environment, Kathmandu consists of an incredible variety of polluting sources including vehicles, dust, wood stoves, and industrial processes. To identify the factors generated by PMF as specific source types we considered the chemical species included in each factor and supported this classification by using the temporal and spatial distribution of emissions. Our study was unique in this part of our analysis as our hourly collection of speciation data enabled us to see how the expression of each individual factor varied over the course of a day: a perspective not available to previous studies using daily measurements. The model was initially run three times, set to construct four, five, and six factors, respectively. From these initial runs, we concluded that four was the optimal number of factors, as it provided three clearly identifiable factors and only one that was inconclusive.

3.1.1 Factor 1: Brick Kilns

Factor 1 represents coal-burning brick kilns, characterized by an extremely high composition of S, as well as small amounts of Fe, K, and As. The kilns burn coal and release large amounts of sulfur in the form of SO_2 as well as black carbon and elemental carbon. Emissions from coal-burning brick kilns contributed 40% of monthly atmospheric SO_2 in the Kathmandu valley (Zhong et al., 2019). Coal from other parts of South and East Asia has been shown to also contain significant concentrations of As (C. Wang et al., 2018). Nearly two thirds (62.2%) of arsenic found in PM2.5 was contributed by this factor. The small amount of iron contributed by this factor likely comes from the bricks themselves, as iron is present in the clay used as the raw

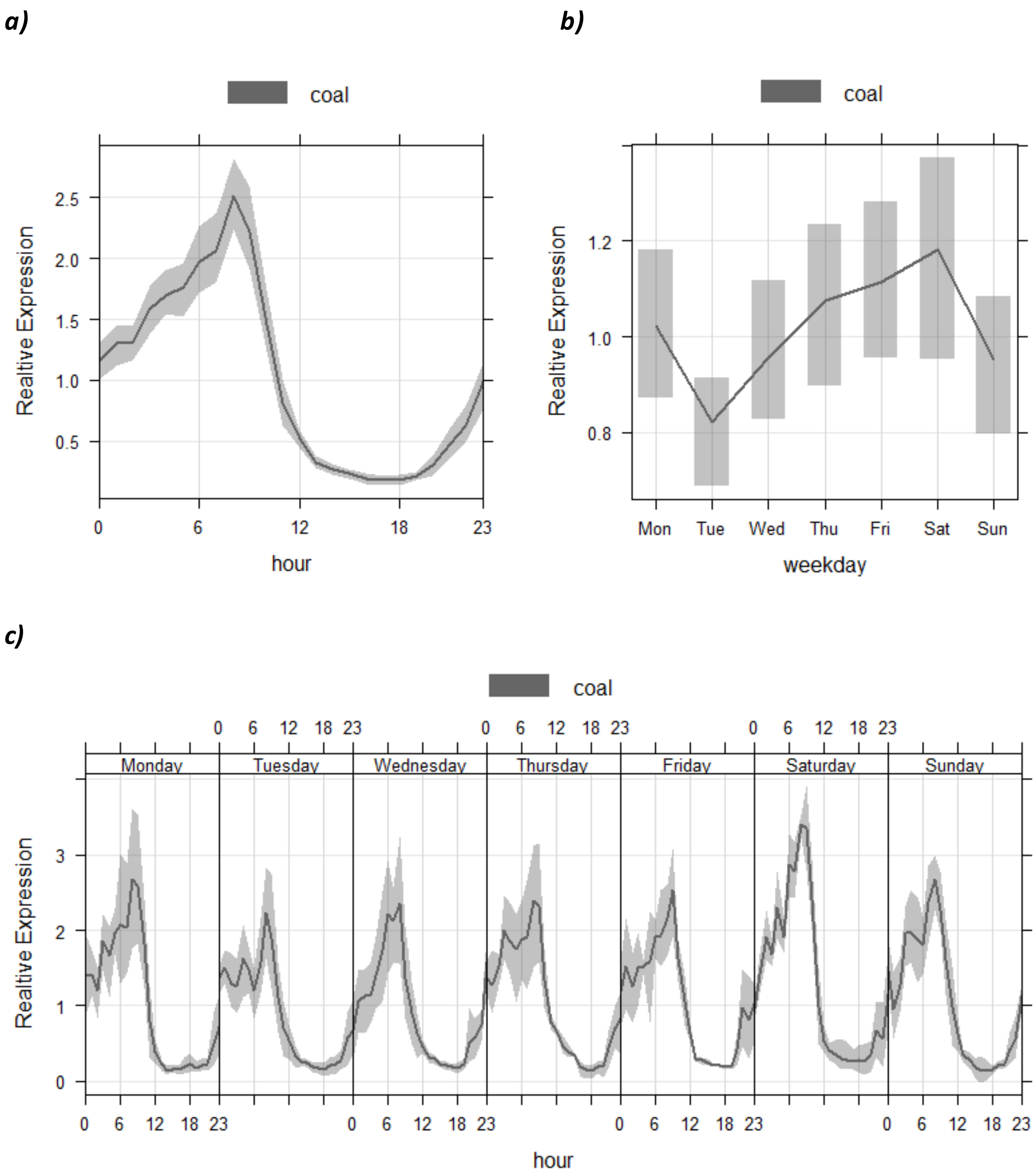

Figure 8: relative expression of factor 1, averaged:
a) hourly, across all sampling days
b) daily, for each day of the week
c) hourly, across the entire week

material for shaping the bricks. The burning of sulfur-containing fuel to bake bricks composed of fine crustal material is most consistent with the composition of this factor.

This is supported by the temporal distribution of factor 1. As shown in Figure 2a, factor 1 increases steadily throughout the course of the day with a sharp drop in concentration mid-morning around 9:00 local time. This same decrease in concentration was observed across all factors and represents the lifting of the atmospheric boundary layer as the air warms and expands. This pattern is consistent with continuously emitting brick kilns.

Figure 5a plots the relative expression of factor 1 with regards to windspeed and wind direction and can be used to approximate the general direction of the main sources of air pollution with regards to the specific sampling location. Factor 1 is most highly expressed when wind blows from the east, northeast, and southeast between 0 and 2 m/s. Figure 3, taken from Zhong et al. (2019), shows the spatial distribution of coal-fired brick kilns in Kathmandu Valley with relation to the sampling location and the Tribhuvan International Airport where meteorological data was collected. The closest cluster of brick kilns to the sampling location shown in figure 3 is located to the east southeast of the site. This corresponds with the highest relative expression of Factor 1, providing further evidence that the source type represented by factor 1 is coal-fired brick kilns.

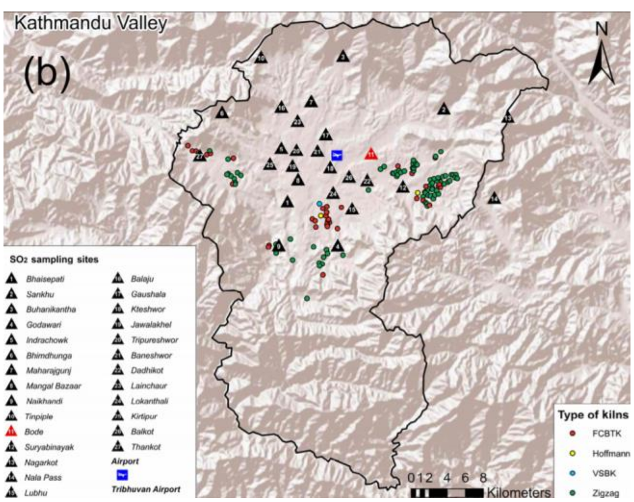

Figure 9: Kathmandu Valley, sampling location marked by red triangle, active kilns marked by circles. Figure taken from Zhong et al., (2019).

3.1.2 Factor 2: Unknown

Factor 2 was unidentifiable as a single source type based on the chemical species included, most notably significant portions of copper, nickel, and zinc. There is no distinct pattern of the temporal distribution of the expression of this factor, as it includes several outliers and large margins of error. It could potentially represent mixed industrial sources, but without further information beyond the 26 metals sampled, accurate identification of this factor is impossible. This factor had a much lower calculated average expression than the others, contributing 2.1%

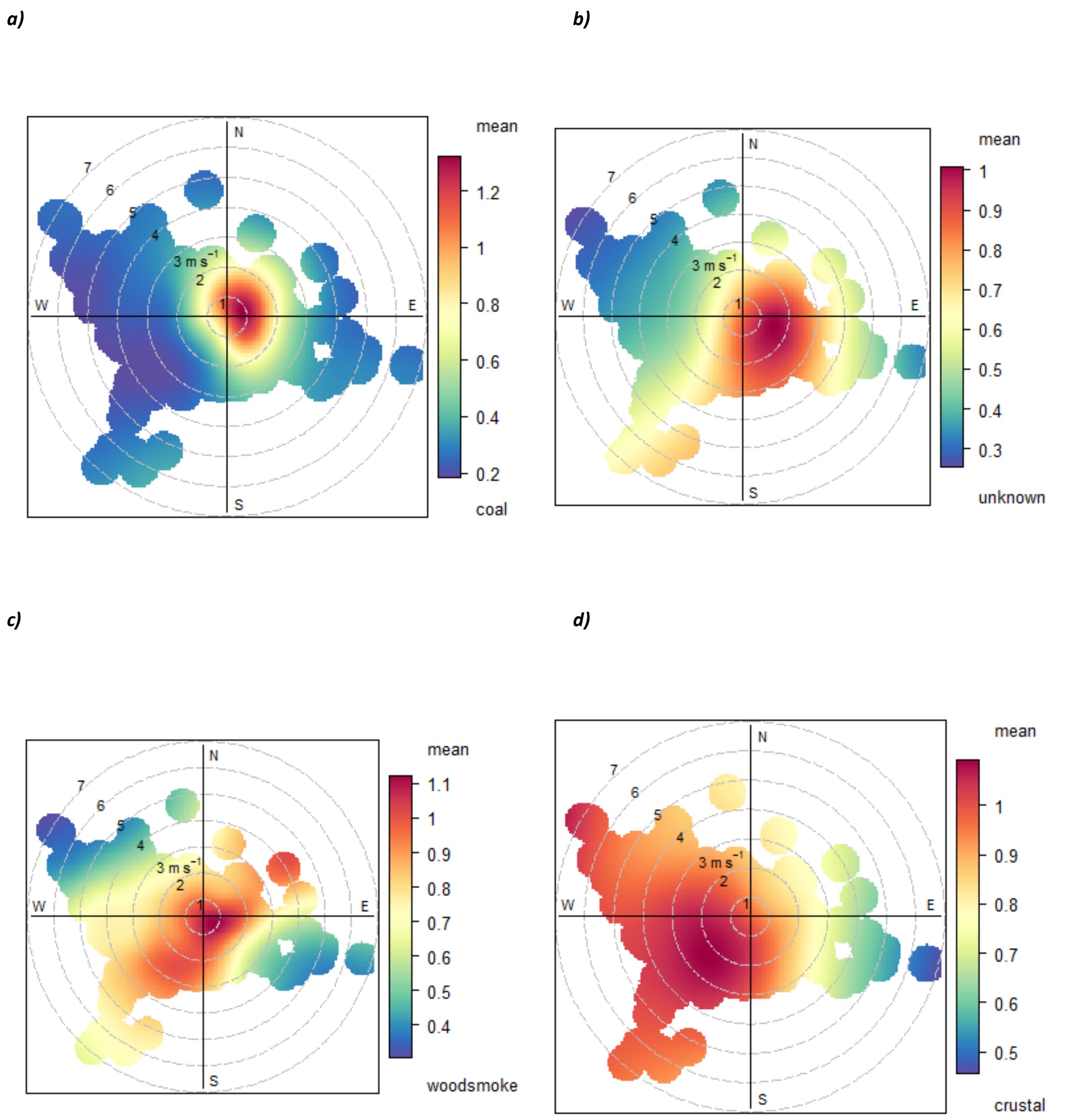

Figure 10: Polar plots for each factor. Each colored circle represents a data point, in which the color represents relative expression shown in the key on the right and the location of the point on the figure represents the weather conditions at the time of sampling. Radial location on the compass rose represents the direction of origin of winds, and distance from the center represents windspeed.

total PM, and so we are confident that the existence of this factor does not compromise the validity of the other 3 factors.

3.1.3 Factor 3: Woodsmoke/Biomass

Factor three is primarily characterized by high concentrations of K, accounting for just over half (56.54%) of all potassium collected. Potassium is a common tracer for emissions from wood-burning stoves and other wood fires and was found in present in significant concentrations in the biomass factor generated by Kim et al. (2015).

The temporal distribution of factor 3 shown in figure 5a is characterized by two large peaks, in the morning and evening, respectively. This is consistent with wood fires used to heat residential homes. The morning peak begins around 5:00 local time when residents are likely waking up and lighting wood stoves to heat their homes for the morning. Expression decreases mid-morning, likely due to the lifting of the boundary layer, and then increases again from about 13:00 until 20:00, after which it decreases through the night until about 5:00. This is consistent with a continued period of activity during the day and into the evening, during which homes need to be heated and cooking takes place, followed by the overnight period during which most home fires are extinguished, causing less woodsmoke to be released.

3.1.4 Factor 4: Crustal Material

The source represented by factor 4 is fine crustal material, characterized by high concentrations of Fe and Ca, as well as the majority of certain trace elements including Ti and Mn. These elements are abundant in the earth's crust and were identified as markers of a crustal factor by Kim et al.(2015). This factor represents fine dust generated by mechanical weathering of the earth's crust made airborne by wind or traffic activity. Our study found a much lower expression of this factor relative to the others than Kim et al.(2015), likely because that study measured PM_{10}, whereas ours measured $PM_{2.5}$. Our study excluded larger particles that are disproportionately generated by mechanical weathering rather than combustion, but the size distribution of these particles does extend into the 2.5µm range, so we do still expect to find some particles of this source type in our analysis.

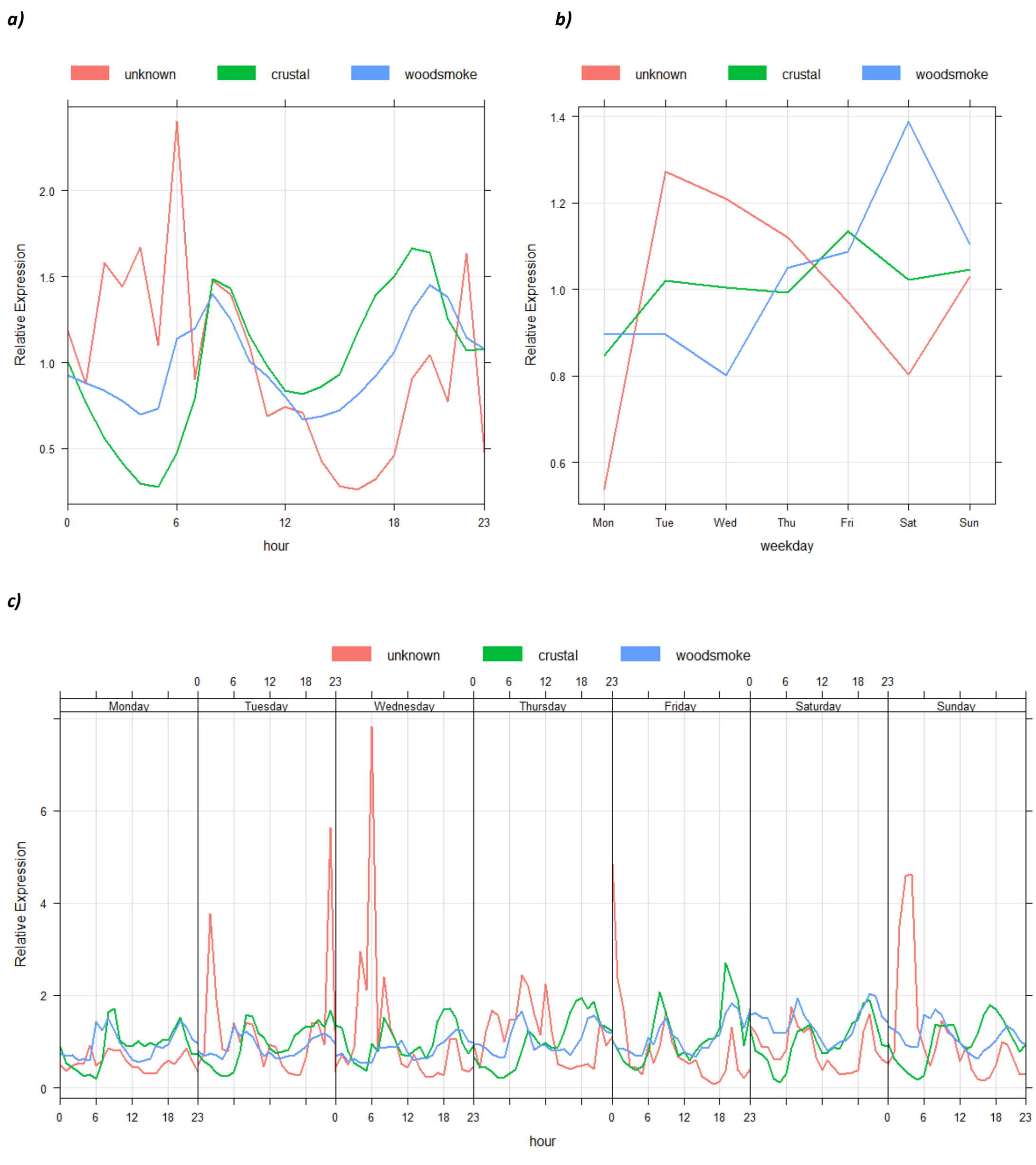

Figure 11: relative expression of factors 2,3, and 4, averaged:
a) hourly, across all sampling days
b) daily, for each day of the week
c) hourly, across the entire week

Specific source types of crustal material in the atmosphere include construction activities, unpaved open areas, and traffic on both paved and unpaved roads, all of which are generally chemically indistinguishable from each other. Dust generated by traffic likely accounts for much of this factor because many of the roads in the Kathmandu valley are unpaved dirt roads. This is supported by the temporal distribution of factor 4, shown in figure 5a. Expression of factor 4 increases during the morning rush hour from about 06:00 local time until 09:00. Pollution decreases to about average between 09:00 and 13:00 due to the lifting of the atmospheric boundary layer, then continues to increase until about 20:00. This represents the normal daily period of high activity, and the following 10 hours show a decrease consistent with the period of low traffic activity during the night.

Figure 4d shows high expression of factor 4 with gentle winds from any direction, with the highest expression occurring during winds from the west and southwest. From the sampling site, this is the direction of the majority of the urban development of Kathmandu. This result is consistent with a crustal source type, given that the majority of crustal material is generated by vehicles, and this is the area with the highest amount of vehicle traffic.

3.2 Estimated Mortality

The main goal of this research was to estimate the mortality attributable to coal in Nepal, which we accomplished with AirQ+ using both in situ source apportionment data and air quality data from the US embassy. We also adjusted the original IER tables by 1.05 to reflect the additional risk posed by coal and used these to make three estimates of mortality. 1) excess mortality attributable to the increased toxicity of coal; 2) total mortality attributable to coal; 3) and total mortality attributable to $PM_{2.5}$.

To make burden of disease calculations, AirQ+ uses a table of Integrated Exposure Response (IER) functions from the Global Burden of Disease study reflecting the risk of air pollution corresponding to a specific population and health endpoint. These risk values are non-specific for PM2.5 and ignore the origin and chemical composition of the pollutant. According to Thurston et al., coal PM has a hazard ratio for IHD equal to 1.05 (95%CI: 1.02, 1.08) (Thurston et al., 2016).

To achieve a baseline of our current understanding of PM health effects, we ran AirQ+ using the original IER and PM data taken from the US Embassy and calculated 9673 estimated deaths, with a low estimate of 3804 and a high estimate of 16614. We then ran AirQ+ a second time, using the modified coal IER and the mean PM concentration from the winter months during which sampling took place. These conditions represent an imaginary scenario in which PM is 100% coal and remains at their highest levels year-round. Taking the difference between our baseline calculations and this worst-case scenario gave a crude understanding of the excess mortality attributable to the increased toxicity of coal, but not one representative of realistic conditions.

To apply this to real-life we first had to adjust for the actual contribution of coal to total PM in relation to other sources. Using our PMF data, we were able to estimate 56.35% of total $PM_{2.5}$ was attributable to coal. We factored our estimate of crude excess death by this value to reflect excess death given the actual relative PM contribution of coal, but with no annual variation.

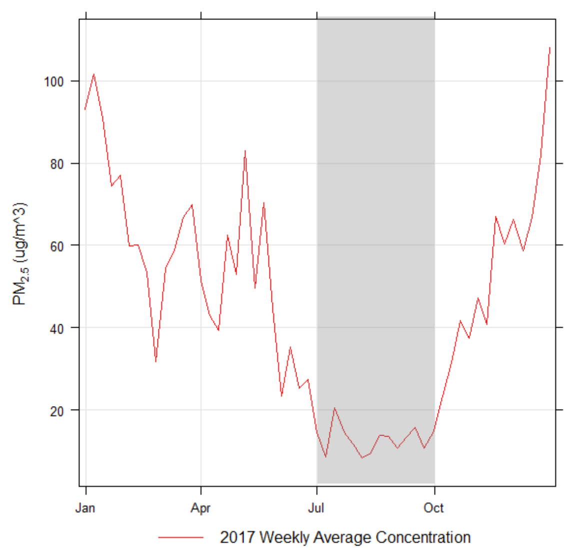

Figure 12: Weekly average concentration of $PM_{2.5}$ measured at the US Embassy in Kathmandu. Months of seasonal non-operation of brick kilns represented by shaded portion.

Weekly average concentration of $PM_{2.5}$ varies greatly throughout the year, reaching lowest concentrations between July and October, pictured in figure 6. To account for annual variation of both total PM2.5 concentration and brick kiln activity, we divided the year into four 3-month seasons and calculated the average PM2.5 concentration during each of these seasons using the full year of embassy data. Sampling occurred during the winter season (January-March) during which the average PM2.5 concentration was the highest (70.52 $\mu g/m^3$), so all health effects calculations until this point were only reflective of those conditions. The current estimate of excess deaths reflects a year of winter conditions, so we divided by 4 to reflect the actual expected number of excess deaths that occur during the winter season. We then factored this value by the average concentrations in the spring (April-June, 46.87 $\mu g/m^3$) and fall (October-December, 51.69 $\mu g/m^3$). Brick kilns in Nepal operate seasonally, with brick production beginning in September/October and ending in June (Joshi et al., 2014). Most registered brick kilns are not in operation during the monsoon season over the summer, the months shaded in figure 6, and therefore are not contributing to adverse health outcomes at that time. For this reason, we do not consider excess death occurring due to air pollution in the summer attributable to brick kiln emissions, and that season is excluded from our health outcome analysis.

With our novel approach, we compute that there were 416 (L: 397, H: 1050) excess deaths that occurred in the winter, spring, and fall, and which would otherwise not be counted without adjusting for the mortality attributable to the increased toxicity of coal pollution. This is also equal to the expected number of lives that would be saved if coal-burning kilns were replaced with similarly-emitting, non-coal kilns. Moreover, these are deaths that would not have been attributed to $PM_{2.5}$ without the information presented in this study, highlighting the importance of source apportionment when conducting risk assessments. By adding these excess deaths to the original baseline mortality estimate, we get a new estimate of 10070 deaths per year from IHD attributable to $PM_{2.5}$. This represents a 4% increase in estimated mortality when accounting for the toxicity of coal.

The total number of deaths attributable to coal was more straightforward, requiring only a seasonal adjustment after the initial AirQ+ calculations. Using the average concentration of the coal factor constructed in PMF and using the modified coal IER, AirQ+ returned a mid-estimate of 4,486.78 cases per year of mortality from IHD attributable to coal PM, reflecting the winter conditions during which sampling took place. After accounting for seasonal changes in PM concentration and brick kiln activity, we reached a final estimate of 2,689 deaths per year attributable to coal emissions in Nepal.

CHAPTER 4

DISCUSSION

4.1 Significance

Our approach combines high-quality pollution data with two types of modeling, each of these informing the next and allowing for a truly unique analysis that would be otherwise impossible. The data collected as a part of this study is the first of its kind; no previous study has conducted source apportionment using hourly time-resolved data in Nepal. This increased resolution allows for more robust source apportionment and analysis at an hourly scale, making this study the first to do so. By using the resulting source apportionment data to inform our impact assessment, we have been able to produce what we expect to be the most accurate assessment of the impact of coal emissions in Nepal to date.

By conducting source apportionment using hourly time-resolved data, we have been able to identify daily patterns of expression of several factors. For the coal factor, the identification of this pattern provides citizens of Nepal with valuable information with which they can make better informed decisions about their daily activity in order to better protect their health. We found the period from noon to 8pm local time to be the daily period of lowest expression for the coal factor, making this the period with not only the lowest concentration of PM, but also presumably the lowest toxicity. By limiting outdoor activity or utilizing PPE outside of these hours, Nepali citizens could minimize their risk.

The benefit of identifying and quantifying specific industrial sources lies in the power it provides governments to more effectively regulate that industry to protect the health of the population. In this specific instance, our study lays the groundwork for a more effective costbenefit analysis to assess the feasibility of implicating new and different environmental policy in Nepal.

Brick kilns, while not an official sector of the Nepalese economy, are the second highest source of particulate matter in Nepal. The industry has some regulations in place, but the regulations are neither comprehensive nor strictly enforced. Additionally, unregistered brick kilns continue to operate unchecked and unaccounted for. Our study has identified a 4% increase in expected mortality that has direct consequences for the people and government of Nepal given the significance of the industry responsible.

On the national scale, one solution that would likely result in less death attributable to the brick industry would be a shift away from coal to a different fuel stock. A 100% transition from coal to another similarly emitting source such as wood/biomass should see around 400 fewer deaths from heart disease per year in the country of Nepal. A shift from coal to a non-PM-emitting source like natural gas (CH_4) would likely result in upward of 2500 less deaths from IHD per year. While a 100% transition for the industry would be a long and expensive process, these results illustrate the real-life benefits of such an undertaking.

4.2 Limitations and Further research

One major limitation of this study is the lack of a transportation signal. Our analysis focused exclusively on metal components of aerosol, and did not evaluate any gasses such as NO_x, an important indicator for the transportation signal. A 2015 study estimated that the transportation sector contributed 80 tons per month, or 16.7% of total monthly NO_x emission in Kathmandu (Janssens-Maenhout et al., 2015). Zhong et al. found this to be a vast underestimation; using an updated model in 2019, they estimated vehicle emissions at 6152 tons per month, or 93.7% of total monthly NO_x. Additionally, they estimated transportation to be responsible for roughly 60% of total monthly $PM_{2.5}$ emissions (Zhong et al., 2019). Comparing this to our results, we expect that a fraction of transportation emissions were included in the coal signal. Future work should broaden the range of pollutants sampled to include gaseous pollutants such as NO_x, SO_2, and VOC's. This would allow for the identification of a transportation factor, and provide a better understanding of the pollution in Kathmandu.

Sampling additional elements would also allow researchers to quantify the full volume of each factor. We believe we have underestimated the actual concentration of each factor as a result of not including all elements in our analysis. This is especially true for the coal factor, which we would expect to include high concentrations of black carbon and carbonaceous ions.

The presence of other coal-burning activities besides brick manufacturing could also have caused us to overestimate the effects of brick kilns. While precise data on miscellaneous coal usage is not available, World Bank reports that as of 2016 only 28% of Nepali Citizens have access to clean cooking technologies, and other fuel sources like kerosene, biomass, and coal are common alternatives. Our study attributed 10-20% more sulfur emissions to brick kilns than previous studies, so it is likely that some of these other sources were included. (Kumar Pariyar et al., 2013; Zhong et al., 2019). Diesel fuel can also be a source of sulfur in $PM_{2.5}$, so it is likely that a fraction of a transportation signal could have been included as well. Inclusion of these additional factors in our coal factor would cause us to overestimate the effects of the coal factor.

Emissions from non-local courses could also have captured in our sampling and introduce some error into our findings. Nepal sits in between the heavily industrialized nations of India and China, and while long range transport of emissions from China is unlikely due to the Tibetan Plateau and general direction of weather patterns at that latitude, emissions from India and Pakistan could much more easily be transported across the relatively flat Indo-Gangetic Plain.

Another limitation of this study is our lack of speciation data outside of the winter months. Because sampling only occurred January through March, we had to assume the relative contribution of each factor to the total PM concentration remained constant. Because brick kiln activity is near-constant through the dry season (October - June), while the overall average PM concentration is lower in the fall and spring, we expect that, during these seasons, the coal factor may have a higher relative expression than during the winter. This would cause an underestimation of health effects in our research, and we would expect the real value to be higher than our estimate.

Additional uncertainty surrounding the relative toxicity of coal emissions are introduced because there is little data on the relative toxicity of other source types found in Nepal, and it is unclear exactly how a pollutant with a risk ratio less than 1.0 would affect these findings. We believe that treating a source type with lower-than-average toxicity as though it were average would cause us to overestimate the total number of deaths attributable to non-coal $PM_{2.5}$ but underestimate the number of deaths attributable to coal $PM_{2.5}$. It is unclear how the total number of deaths attributable to $PM_{2.5}$ would be affected.

We believe our approach could be implemented effectively in other areas of the world where PM is dominated by one or two particular source types. For example, using source apportionment and source-specific mortality response data could help researchers better understand the health impacts of wildfires in the American West, a topic of growing concern and relevance with climate change.

CHAPTER 5

CONCLUSION

Our study highlights the importance of source apportionment and relative toxicity when assessing health impacts of ambient PM2.5; we discovered a 4% increase in mortality that would be otherwise overlooked. If this kind of meaningful effect is observed in a relatively small sample location like Kathmandu, we expect other parts of the world that are more polluted and more populated would benefit tremendously from this approach.

Our methods also have the potential to improve causality precision in air pollution epidemiology by reducing exposure uncertainty and exposure misclassification. By combining these commonly-used tools, we have been able to isolate the effects of a specific fraction of air pollution in a location and make assumptions about the specific source type responsible.

CHAPTER 6

ADDITIONAL RESOURCES

These tables provide additional information regarding specific aspects of our research. Table 2 provides the species sampled for source apportionment, and table 3 shows the stratified health data used in AirQ+.

Sampled Species			
Sulfur	Chromium	Zinc	Cadmium
Potassium	Manganese	Arsenic	Tin
Calcium	Iron	Selenium	Antimony
Scandium	Cobalt	Bromine	Barium
Titanium	Nickel	Rubidium	Lead
Vanadium	Copper	Strontium	

Table 2: elements sampled using an XACT 620

Table 2
Deaths from CVDs in Nepal by age groups and gender in 2017.

Age groups (yrs)	Death per 100,000 (95% UI)			Percentage of total deaths (95% UI)		
	Both	Male	Female	Both	Male	Female
1–4	0.4 (0.7–0.2)	0.6 (1.0–0.3)	0.3 (0.5–0.1)	0.4 (0.7–0.2)	0.6 (0.9–0.3)	0.3 (0.5–0.1)
5–9	0.3 (0.5–0.2)	0.4 (0.6–0.2)	0.3 (0.5–0.2)	0.7 (1.0–0.4)	0.7 (1.0–0.4)	0.7 (1.0–0.4)
10–14	0.7 (0.9–0.4)	0.8 (1.2–0.5)	0.5 (0.8–0.3)	1.6 (2.2–1.1)	1.7 (2.5–1.1)	1.4 (2.1–0.8)
15–19	3.9 (5.1–2.7)	5.8 (8.2–3.9)	2.0 (2.8–1.3)	4.8 (6.2–3.4)	5.7 (7.9–3.9)	3.2 (4.5–2.2)
20–24	7.2 (9.8–4.9)	11.7 (17.2–7.4)	3.2 (4.6–2.2)	6.6 (8.8–4.6)	8.2 (11.3–5.3)	4.1 (5.7–2.9)
25–29	9.6 (13.5–5.1)	15.4 (23.4–7.0)	5.1 (7.2–2.8)	8.4 (11.2–4.4)	10.9 (7.5–3.0)	5.4 (7.5–3.0)
30–34	15.8 (22.9–4.7)	23.0 (37.2–3.0)	10.3 (14.7–4.8)	10.8 (14.8–3.3)	13.8 (20.9–1.9)	7.9 (10.9–3.7)
35–39	28.3 (39.6–10.3)	38.4 (59.7–5.0)	20.4 (29.0–12.3)	13.7 (18.5–5.0)	16.9 (25.1–2.1)	10.8 (14.3–7)
40–44	63.0 (82.6–34.4)	83.6 (121.4–26.9)	46.1 (63.1–34.4)	20.4 (25.2–11.3)	24.2 (31.9–8.7)	16.4 (20.5–12.3)
45–49	124.1 (156.8–86.1)	168.9 (234.5–102.2)	84.2 (110.8–60.5)	25.9 (30.6–19.4)	30.7 (37.6–20.3)	20.3 (24.7–16.2)
50–54	237.0 (287.3–187.9)	338.2 (432.2–244.1)	140.7 (186.1–104.4)	31.0 (35.1–26.1)	37.3 (42.9–30.0)	22.3 (27.1–18.0)
55–59	405.1 (475.8–328.6)	578.4 (703.4–440.2)	238.7 (312.6–178.0)	34.1 (37.8–30.2)	39.3 (44.0–34.1)	26.1 (30.8–20.9)
60–64	653.7 (756.1–539.5)	953.1 (1131.4–738.4)	374.0 (476.8–284.5)	34.9 (38.7–30.9)	40.4 (45.1–35.5)	26.5 (31.5–21.3)
65–69	1038.6 (1190.2–878.5)	1503.6 (1742.5–1199.7)	617.0 (797.6–474.5)	36.2 (40.3–32.5)	41.6 (46.2–37.0)	28.2 (33.8–22.8)
70–74	1557.5 (1782.2–1323.2)	2254.4 (2570.3–1822.2)	892.7 (1148.4–661.9)	34.6 (38.3–30.7)	40.7 (45.1–36.0)	25.4 (31.5–19.4)
75–79	2442.9 (2771.2–2118.6)	3407.0 (3854.0–2807.5)	1537.0 (1964.4–1135.3)	35.0 (38.8–31.4)	40.5 (45.0–36.0)	27.2 (33.5–20.8)
80 above	4274.7 (4807.6–3777.9)	5938.5 (6636.2–5075.9)	2994.7 (3760.7–2345.3)	30.6 (34.2–27.7)	37.3 (41.0–33.4)	24.0 (29.4–19.6)
Total	260.8 (292.3–227.6)	372.0 (414.6–309.7)	165.4 (200.8–134.6)	27.0 (29.5–24.4)	32.1 (34.8–29.2)	20.3 (23.4–17.4)

Table 3: Stratified mortality data from Nepal for CVD in 2017, from Bhattarai et al., (2020).

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