## **Attribution of PM2.5 Health Impacts in Asia-Pacific**

by

#### Kingshuk Dasadhikari

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Signature of Author: …………………………………………………………………………………………………………………… Department of Aeronautics and Astronautics

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Certified by: ……………………………………………………………………………………………………………………………….. Steven R.H. Barrett Associate Professor of Aeronautics and Astronautics Thesis Supervisor

Accepted by: ………………………………………………………………………………………………………………………………. Hamsa Balakrishnan Associate Professor of Aeronautics and Astronautics Chair, Graduate Program Committee

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#### **Abstract**

Asia-Pacific anthropogenic emissions have changed rapidly in recent years due to industrialization, increasing mobility, and emissions controls. Although these changes have altered the region's burden of premature mortalities due to ambient fine particulate matter  $(PM<sub>2.5</sub>)$ , the contribution of each sector and effectiveness of different policy measures has not yet been quantified. Such data would inform future decision-making on both policy effectiveness and the relative importance of controlling emissions from different sectors. This study estimates changes in regional anthropogenic emissions by industrial sector between 2010 and 2015, based on sector-level activity indicators and enacted emission controls. These factors are applied to an existing high-resolution emissions inventory for 2010 to estimate emissions up to 2015. Using a chemical transport model, the effects of changes in each sector's contribution to total  $PM_{2.5}$ driven premature mortalities are calculated for 2010 – 2015, in addition to the total contribution of each sector to premature mortality in 2015. 2,000,000 (95% CI: 1,740,000-2,260,000) annual global PM2.5-driven premature mortalities are attributed to Asia-Pacific anthropogenic sectoral emissions in 2015. The agricultural, industrial, and residential sectors constitute the top three sources of these total impacts. Between 2010 and 2015, sustained economic and activity growth, particularly in South and Southeast Asia, have led to  $129,000$  (95% CI:  $106,000-166,000$ ) additional annual premature mortalities, primarily across India, Indonesia, and Bangladesh. The energy and industrial sectors, in particular, cause 38,000 and 45,000 additional annual premature mortalities across these three countries respectively. Simultaneously, falling activity rates in other countries due to structural changes such as electrification of railroads, as well as newly introduced abatement measures over this period, including China's Action Plan on the Prevention and Control of Air Pollution as well as region-wide adoption of Euro IV/V/VI-compliant road vehicle emission and fuel quality standards have led to a total reduction of 95,000 (95% CI: 76,000-129,000) annual premature mortalities, primarily across East Asia, including China and Japan. These opposing drivers result in a net change of an additional 34,000 (95% CI: 23,00047,000) PM2.5-driven annual premature mortalities between 2010 and 2015 due to Asia-Pacific anthropogenic emissions.

Thesis Supervisor: Steven R. H. Barrett Title: Associate Professor of Aeronautics and Astronautics THIS PAGE INTENTIONALLY LEFT BLANK

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### **1 Introduction**

Anthropogenic emissions, from activities such as combustion and agriculture, directly and indirectly affect surface air quality, leading to negative impacts on human health such as increased mortality and morbidity. Past research has estimated more than 6.4 million premature mortalities annually [10] due to human exposure to air pollution in recent years, exceeding the burden of other prominent global health threats such as malaria [3]. Of these mortalities, 4.2 million are attributed to ambient particulate matter of a diameter of less than 2.5  $\mu$ m (PM<sub>2.5</sub>).

Over 50% of these mortalities occur in the Asia-Pacific region, with over 2 million premature mortalities in China and India attributed annually to PM2.5 exposure [10]. Regional emissions of PM<sub>2.5</sub>, including primary PM<sub>2.5</sub> and PM<sub>2.5</sub> precursors, are estimated to have increased by 20-50% from 2000 to 2010 [11] (**Figure 1-1**). Over the same period, the regional population grew significantly while becoming increasingly concentrated in polluted urban centers [45]. However, in some areas these factors have begun to be countered by emissions controls measures, such as China's Action Plan on the Prevention and Control of Air Pollution [9]. Although most significant for local air quality, pollution reductions from such measures are transported beyond national borders, with possible benefits over regional and even global scales. The pattern and quantity of benefits depends on the affected sector and species, the local and regional meteorology, and the background atmospheric composition.

In this context, there is a need to better understand the trends of anthropogenic emissions and air pollution in this region in order to evaluate the effectiveness of current mitigation strategies as well as to improve future strategies to better counteract the regional and global burden of premature mortalities arising due to anthropogenic emissions from this region. A

greater understanding of the causes of air pollution in this region, including the key sectoral drivers and effect of existing emissions controls on sources as well as the spatial distribution and temporal evolution of regional and global impacts, is necessary to inform the development of such strategies.



Figure 1-1: Growth in emissions of primary PM<sub>2.5</sub> and PM<sub>2.5</sub> precursors in the Asia-Pacific region between 2000 and 2010 in Tg/year. Data taken from the Emission Database for Global Atmospheric Research (EDGAR) [11].

This study addresses this problem in two stages. In the first stage, we develop a sectoral anthropogenic emissions inventory for the Asia-Pacific region that spans the 2010-2015 period, fully covering the  $12<sup>th</sup>$  Five-Year Plan of the People's Republic of China. Existing emissions inventories for the region were either produced prior to the period in question (e.g. Streets-2000 [42], INTEX-B [55], MIX [29], and EDGAR [11]) or lack the necessary sectoral, spatial, or temporal resolution (e.g. ECLIPSE [2, 23] and CEDS [20]). Furthermore, although the MEIC regional emissions inventory [29, 31, 55, 57] has been continuously updated up to 2015, these updates

have only been estimated for Chinese emissions, furnishing no updated emissions data for the other countries in this region.

To isolate the effect of specific fuel use and emissions policies, we develop sector-specific growth factors for the 2010-2015 period that incorporate sector-level activity indicators, published emissions standards, and information on emissions trends from the existing regional MIX 2010 and MEIC 2015 inventories [29, 31, 55, 57]. We apply these data to the existing EDGAR emissions inventory calculated for 2010 [11]. This composite approach results in an inventory with fine sectoral resolution, emissions projections based on reported rather than projected activity indicators and emissions controls, and applicability over the time period of interest (2010- 2015). The updated emissions inventory directly characterizes both the temporal evolution of regional anthropogenic emissions in recent years as well as the effect of existing emissions controls at a sectoral level.

In the second stage, this inventory is incorporated into the GEOS-Chem chemical transport model (CTM) and used to simulate years 2010 and 2015. This allows us to characterize both the regional and global spatial distribution and temporal evolution in air quality impacts by sector due to the modeled change in regional sectoral emissions. Specifically, we estimate the net change in regional and global mortality due to Asia-Pacific anthropogenic sectoral emissions between 2010 and 2015, the breakdown of relative contribution of changes in emissions compared to demographic factors to this net change, and the effect of growth and emissions controls in each of eight sectors on mortality in each of 11 countries/country groupings in the region, as well on mortalities exported to other global regions.

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### **2 Methods**

This section describes the methods applied for both the generation of updated emissions up to 2015 for the Asia-Pacific region, as well as for air quality modeling and health impact quantification using the 2010 and 2015 emissions to study the sectoral contributions to total global and regional mortalities in 2015 stemming from this region, and to the change in such mortalities between 2010 and 2015.

### **2.1 Emissions**

The emissions for 2015 used in this study are generated by applying a scaling method to EDGAR 4.3.1 baseline sectoral emissions for 2010. The EDGAR baseline emissions, jointly compiled by the European Commission Directorate General Joint Research Center (JRC) and the Netherlands Environmental Assessment Agency (PBL) [11] are available as a comprehensive gridded dataset of global emissions for 14 different economic sectors described in **Table 2.1** and six different chemical species, including ammonia (NH<sub>3</sub>), carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>), speciated non-methane volatile organic carbon (NMVOCs), primary PM2.5 (including speciated BC and OC), and sulfur dioxide  $(SO<sub>2</sub>)$ .

The baseline EDGAR v4.3.1 global anthropogenic emissions inventory are computed using a consistent, technology-based emission factor approach [11]. This provides average, monthly, sector-specific emissions of both air pollutants and greenhouse gases spanning a 40-year time series from 1970 to 2010, gridded spatially at a resolution of  $0.1^{\circ} \times 0.1^{\circ}$  degrees. EDGAR emissions have been widely applied in air quality modeling, including as the standard emissions inventory for CTMs such as GEOS-Chem, and have also been incorporated into the development of other emissions inventories, such as HTAP, ECLIPSE, and CEDS global inventories and the REAS pre-2010 inventories for Asia [2, 20, 22, 23, 25].

<b>Sector</b>	<b>Description</b>		
<b>Agriculture</b>	Fertilizer use, animal husbandry, rice cultivation, waste burning		
<b>Aviation</b>	Take-off and landing, climb and descent, cruise, and supersonic		
<b>Energy</b>	Power and heat generation (including cogeneration/CHP)		
<b>Manufacturing</b>	Manufacturing		
<b>Fossil Fuel Fires</b>	Coal, oil, and gas fires		
<b>Industrial Processes</b>	Cement, lime, mineral, metal, food, and paper/pulp production		
<b>Fuel Exploitation</b>	Oil and gas extraction		
<b>Residential</b>	Buildings/human habitation		
<b>Oil Refineries</b>	Oil refineries		
<b>Shipping</b>	Domestic and international shipping		
<b>Waste Processing</b>	Emissions from waste incineration		
<b>Rail Transportation</b>	Emissions from rail vehicles and other ground transportation		
<b>Energy Transformation</b>	Fuel processing (fugitive emissions)		
<b>Road Transportation</b>	Emissions from road vehicles		

**Table 2.1:** Sectors covered by the EDGAR 4.3.1 emissions inventory [11]

Out of 208 countries with sectoral emissions considered by EDGAR, 25 from the Asia-Pacific region are selected for update up to 2015. Combined, this selection of countries contains a population of 3.4-3.7 billion people (as of 2010 and 2015 respectively). A list and map of these countries, spanning East Asia, Southeast Asia, and most of the Indian subcontinent/South Asia (with the exception of Afghanistan and Pakistan) is shown in **Figure 2-1**.

All chemical species constituting the EDGAR emissions inventory have direct or secondary impacts on aerosol chemistry, and therefore are relevant to this study. Species with direct impacts include both primary  $PM_{2.5}$  (including BC and OC), and secondary  $PM_{2.5}$  precursors such as NH<sub>3</sub>, NO<sub>x</sub>, and SO<sub>2</sub>. However, for the purpose of this study, only 12 out of the 14 sectors in EDGAR 4.3.1 were considered, neglecting changes in aviation emissions and fossil fuel fires, which were found to have negligible emissions and little variance over time in this region. Furthermore, while the 12 sectors are treated distinctly for the purposes of emission modeling up to 2015, for impact computation as presented in the Results section, the manufacturing and industrial process sectors are combined into a single "industrial" sector. Similarly, the fuel exploitation, oil refinery, and fuel transformation sectors are combined into a single "fuel processing" sector, and the shipping and waste processing sectors are combined into "other", giving eight separate sectoral groupings in total.



**Figure 2-1:** Asia-Pacific countries considered in this study; countries are colored according to subregion of applied road vehicle emissions factors (section **2.1.3**).

### **2.1.1 Activity-driven scaling**

In order to model changes in regional sectoral emissions for the 26 countries considered between 2010 and 2015, country-level information concerning variations of human activity with respect to each sector (e.g. fuel use for most combustion-driven sectors) as well as country-level changes in emissions standards and other abatement measures are combined into a scaling model. The general form of this model is presented in equation **(2.1)**

$$
E(c, n, s, m, y) = \left(\sum_{i=1}^{i_{\text{ind}}(c)} \alpha_i(c, n, s) r_i(c, n, s, m, y) \frac{I_i(c, n, y)}{I_i(c, n, s, 0)}\right) E(c, n, s, m, 2010)
$$
 (2.1)

where:

 $E$  is emissions from a particular sector, country, species, and time

 $c$  is the index for the emissions sector

 $n$  is the index for the emitting country

 $s$  is the index for the emitted species

 $m$  is the index for the month of the year (1-12)

 $y$  is the index for the year (2010-2015)

 $i_{\text{ind}}$  is the number of activity indicators pertinent to each sector

 $\alpha$  is an apportioning factor weighting each activity indicator of a sector

 $r$  is an abatement factor for each activity indicator of a sector

 $I$  is each activity indicator for a sector

For sectors with unique activity indicators and no modeled abatement measures, equation **(2.2)** reduces to

$$
E(c, n, s, m, y) = \frac{I(c, n, y)}{I(c, n, 2010)} E(c, n, s, m, 2010)
$$
\n(2.2)

For sectors with more complex human activity, dynamics involving multiple activity indicators and/or existing abatement measures such as emissions controls, apportioning and abatement factors are derived individually to quantify the effect of each indicator on total change in emissions, as detailed later in equations **(2.3)** and **(2.4)**. Over the modeled five-year period, we assume that the spatial distribution of emissions for each sector in each country remains constant, with a small number of exceptions (see section 2.1.5).

### **2.1.2 Sector-specific activity indicators**



**Table 2.2:** List of activity indicators by sector

Activity indicators for each EDGAR sector considered for this study are compiled from a variety of sources. For most combustion-driven sectors, fuel use indicators are sourced from the United Nations Statistics Division Energy Statistics Database (UNSD-ESD) [46]. This database details each country's energy flows and balances on an annual basis, incorporating usage statistics of over 50 different industrial substances and fuels, including a variety of oils and types of coal, by more than 40 different activities spanning all combustion-driven sectors included in EDGAR. In addition, the database also provides direct activity statistic for some sectors, such as refinery throughput for oil refineries, as well as oil and gas production for the fuel exploitation and energy transformation sectors. The UNSD-ESD database provides higher resolution of data in terms of activity drivers and fuels than alternative sources such as IEA Energy statistics used to generate the CEDS global emissions inventory [20].

For sectors that are not combustion-driven, or whose activities are not well characterized in the UNSD-ESD database, such as agriculture, industrial process emissions, and waste processing, alternative activity indicators are used. All activity indicators for the agricultural sector are from statistics and indices provided by the United Nations Food and Agricultural Organization (FAO) [14]. Activity indicators for industrial process emissions are derived from three sources, namely the U.S. Geological Survey (USGS) for production statistics of aggregates, metals, and minerals [50]; BASF corporation for regional growth statistics of chemical production [4]; and FAO statistics for food processing and paper production [14]. Waste processing emissions are scaled with urban population data obtained from the World Bank Databank, originally sourced from the UN Population Division (UNPD) [45]. **Table 2.2** summarizes specific activity indicators and their sources for both combustion-driven and other sectors.

#### **2.1.3 Treatment of sectors with multiple indicators**

Of the sectors listed in **Table 2.1**, only three (oil refineries, waste processing, and rail transportation), are driven by a single activity indicator. The remaining nine are driven by multiple activity indicators. For such sectors, it is necessary to compute apportioning factors to determine the fraction of 2010 emissions attributable to each activity indicator in order to scale emissions using equation **(2.1)**. For the agriculture and industrial process sectors, a breakdown of total sectoral emissions by sub-activity is provided by EDGAR emissions summary files, from which apportioning factors for each chemical species can be computed directly. For the remaining

sectors, apportioning factors must be computed on the basis of sector-specific emissions factors and 2010 activity indicators. These factors are applied into the single factor

$$
\alpha_i(c, n, s) = \frac{f_i(c, n, s, m, 2010)I_i(c, n, 2010)}{\left(\sum_{i=1}^{i_{\text{ind}}(c)} f_i(c, n, s, m, 2010)I_i(c, n, 2010)\right)}
$$
(2.3)

where  $f_i$  is the sector-specific emission-factor for each activity indicator and species and all other variables are consistent with equation **(2.1)**

<b>Sector</b>	<b>Emissions Factors For:</b>	Source(s)	
<b>Energy</b>	Anthracite*†, Bituminous Coal*†, Fuel Oil^, Lignite*†, Natural Gas^ combustion in power/heat plants	^ U.S. EPA AP-42 [47] $\dagger$ Xu et al. [53] * Zhao et al. [56]	
<b>Manufacturing</b>	Anthracite, Bagasse, Bituminous Coal, Blast Furnace Gas, Diesel, Fuel Oil, Fuelwood, Kerosene, LPG, Natural Gas combustion by manufacturing facilities	U.S. EPA NEI-2014 [49]	
<b>Fuel Exploitation</b>	Crude Oil, Natural Gas extraction/processing/ transportation	Lewis [27]	
<b>Residential</b>	Anthracite, Bituminous Coal, Diesel, Fuel Oil, Fuelwood, Kerosene, LPG, Natural Gas, Vegetal Waste combustion by commercial facilities/households	<b>U.S. EPA NEI-2014</b>	
<b>Shipping</b>	Diesel, Fuel Oil combustion in marine engines	Winnes & Fridell [52]	
<b>Energy Transform.</b>	Crude Oil, Natural Gas extraction/processing/ transportation	Lewis	
Road Transport.	Diesel, Gasoline combustion in automotive engines (country-specific)	Country-specific emissions standards	

**Table 2.3:** Sources of sector-specific emission factors

Sector-specific emissions factors for all sectors that require this method of apportioning are obtained from multiple sources as detailed in **Table 2.3**. While for most sectors, the emissions factors described in **Table 2.3** apply region-wide, for road transportation, emissions factors are obtained nationally for 6 major countries in this region: China, India, Japan, Indonesia, South Korea, and Thailand, and applied sub-regionally as denoted by the coloring of countries in **Figure 2-1**.

#### **2.1.4 Applied emissions controls**

For sectors that are known to undergo changes in terms of emissions regulations and controls between 2010 and 2015, these are included in the computation of changes in emissions over the five-year period through abatement factors applied to equation **(2.1)**. The derivation of abatement factors for all relevant sectors is calculated as

$$
r_i(c, n, s, m, y) = \frac{f_i(c, n, s, m, y)}{f_i(c, n, s, m, 2010)}
$$
(2.4)

where all variables are consistent with equations **(2.1)** and **(2.3)**

Abatement factors are used for the energy, manufacturing, and residential sectors in China and for the shipping and road transportation sectors region-wide. In the case of the Chinese energy sector, abatement measures are applied for  $NO_{x}$ , primary  $PM_{2.5}$ , and  $SO_{2}$  emissions.  $NO_{x}$ emissions are estimated according to Liu et al. [32], which finds a 60% reduction in the emission factor of NO<sub>x</sub> from Chinese coal-fired plants between 2011 and 2015 as a result of the widespread penetration of selective catalytic reduction technologies. For emission modeling, this reduction is represented as a linear ramp in the  $NO<sub>x</sub>$  emission factor for Chinese coal-fired power plants. Abatement measures are also applied for primary  $PM_{2.5}$  and  $SO_2$  emissions from the Chinese energy, manufacturing, and residential sectors. For these emissions, abatement factors are derived to match trends in the MEIC regional inventory [29, 31, 55, 57], as documented by Zheng et al. [58]. For both species across the three sectors, reductions are observed as a result of China's Action Plan on the Prevention and Control of Air Pollution, in effect since 2013 [9, 58].

For the regional shipping sector, abatement factors are derived for  $SO<sub>2</sub>$  emissions from international shipping in accordance with International Maritime Organization regulations mandating a reduction of fuel sulfur content in international marine bunker fuel from 4.5% to 3.5% from 2012 onwards [41]. A more detailed consideration of emissions controls is applied to the regional road transportation sector, where abatement factors are derived for all emissions species, and for all six major countries in this region whose road vehicle emissions factors are applied sub-regionally. Trends in emissions factors for this sector, including uptake time, are estimated from nationally published vehicle emissions and fuel sulfur standards for both diesel and gasoline.

Although the region-wide shipping and road transportation sectors, and the Chinese energy, manufacturing, and residential sectors are the only ones where emissions controls are explicitly modeled through application of abatement factors, controls that affect overall fuel use are captured in the sector-specific activity indicators. This includes transitions between different types of fuels (e.g. from coal to natural gas in the case of energy emissions in Myanmar) and electrification (e.g. in the case of the railroad sector in China).

#### **2.1.5 Other considerations**

While the speciation of NMVOCs for the updated inventory is provided by EDGAR, no such speciation was provided for PM2.5, other than of black and organic carbon (BC and OC). As a result, the remaining component of  $PM<sub>2.5</sub>$  was segmented into individual species including primary ammonium, nitrates, and sulfates as well as trace metals on the basis of a speciation profile developed by Fu et al. [15] for 24 different combustion sources that cover all 12 EDGAR sectors considered in this study.

Furthermore, although the scaling approach described was applied uniformly across all 12 sectors considered, it was found to be ill suited for modeling changes in emissions from the energy sector for the countries of Cambodia and Sri Lanka as a result of major changes in both fuel consumption patterns and spatial patterns of emissions. This is due to the opening of the first large-scale coal-fired power plants (Sihanoukville I and II plants in Cambodia, and Lakvijaya plant in Sri Lanka) in these countries within the 2010-2015 period. For these unique cases, the

change in emissions as a result of the opening of these plants is modeled by incorporating emissions from coal-fired plants of similar scale from the GPED 2010 Global Power Emissions Database [44].

## **2.2 Air quality modeling**

The GEOS-Chem chemical transport model (CTM) is used to simulate the effect of changes in sectoral emissions from 2010 to 2015 on surface-level concentrations of PM<sub>2.5</sub>. GEOS-Chem uses nested Eulerian grids to simulate atmospheric processes including emissions, chemistry, transport, and deposition, providing hourly estimates of the resulting ground-level concentration of PM<sub>2.5</sub>. First developed by Damian et al. [5], the GEOS-Chem CTM uses the Kinetic PreProcessor chemical solver [12] and the RPMARES aerosol equilibrium model [6] to perform both gas- and aerosol-phase chemistry. For this project, the forward modeling component of GEOS-Chem adjoint v35 [19] is used. This variant of GEOS-Chem couples the conventional forward model capabilities of a CTM with additional inverse capabilities that can be used for inverse modeling, data assimilation, and sensitivity studies. Version 35 of this model incorporates most updates applied up to 2017, and the specific model applied for this study also contains further updates to input emissions, including global 2010 EDGAR 4.3.1 emissions and the Asia-Pacific emissions updated up to 2015.

In this study, the GEOS-Chem CTM is run over a global domain with  $4^{\circ}$  latitude  $\times$  5° longitude resolution to obtain global impacts of modeled sectoral emissions and emissions changes in the Asia-Pacific region. This global simulation is also used to provide boundary conditions to a nested  $0.5^{\circ} \times 0.666^{\circ}$  grid centered on the Asia-Pacific region, providing a high-resolution estimate of regional impacts. The finer regional grid extends between  $11^{\circ}$  S – 55° N latitude and between  $70^{\circ}$  E – 150° E longitude, with boundary conditions specified by the global runs. Although this regional grid excludes a small part of the western Indian state of Gujarat, the population of the excluded portion is less than 0.1% of the total population of India and therefore, has negligible effect on the results of this study. Across all simulations, a variable-vertical-resolution 47-layer hybrid sigma-pressure grid is used, with a model top of 80 km altitude. GEOS-Chem is driven by

meteorological data from the forward-procession output of the GEOS data assimilation system. This data is provided by the NASA Global Modeling and Assimilation Office.

Outside of the Asia-Pacific region, the unmodified 2010 EDGAR emissions inventory is used. Where available, this is superseded by regional emissions inventories such as the U.S. EPA NEI-2011 inventory for the United States of America [48]. Wildfire emissions for each year are provided by the GFED4 wildfire inventory distributed by Oak Ridge National Laboratory [39]. Geogenic emissions such as lightning  $NO<sub>x</sub>$ , dust, and sea-salt aerosols are modeled using the LIS/OTD dataset from NASA's Global Hydrology Resource Center (GHRC) [8], the dust entrainment and deposition (DEAD) mobilization scheme developed by Zender et al. [54], and the sea-salt simulation model developed by Alexander [1] respectively. Background aviation emissions are taken from the Federal Aviation Administration (FAA) Aviation Environmental Design Tool (AEDT) inventories [13].

To analyze the contribution of each sector to the change in global and regional health impacts between 2010 and 2015, a series of 18-month GEOS-Chem runs are conducted. The first 6 months of each run are discarded as a model spin-up period to remove the influence of initial conditions and the remaining 12 months averaged to provide the annual-average concentration of  $PM_{2.5}$ . To quantify the total global and regional population exposure to  $PM_{2.5}$  attributable to sectoral emissions from the Asia-Pacific region in 2015, as well as the change over the 2010 – 2015 period, both global and regional domain runs are first conducted with all Asia-Pacific sectoral emissions set to 2015 values, and eliminated or reverted to 2010 respectively. This is then repeated for each of the 12 sectors considered: two global domain runs and two regional domain runs are conducted with emissions for only that sector completely eliminated for the first pair of global/regional runs and reverted from 2015 values to 2010 values for the second pair.

The difference between annual-average ground-level  $PM_{2.5}$  distributions from runs with all emissions set to 2015 values and runs with individual sectors reverted to 2010 allows computation of each sector's contribution to the change in global/regional  $PM_{2.5}$ -driven mortalities between 2010 and 2015. Applying the same method to the runs with emissions eliminated by sector allows computation of each sector's contribution to total  $PM_{2.5}$ -driven mortalities attributable to 2015 Asia-Pacific emissions.

### **2.3 Health impact assessment**

To compute premature mortalities attributable to each economic sector from the ground-level PM2.5 distributions obtained from GEOS-Chem, we use the Integrated Exposure Response (IER) function developed by [7]. The IER is used to estimate the effect of increased PM<sub>2.5</sub> exposure on mortality rates from chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), lower respiratory infections (LRI), lung cancer, and cerebrovascular disease (strokes). Parameters for each IER function are adopted from the latest WHO Global Burden of Disease (GBD) 2016 study [16]. The mean and 95% confidence intervals of IERs for each of the five health endpoints are presented in **Figure A-1**. While a single curve is used to characterize relative risk of COPD, LRI, and lung cancer at all ages, two sets of 15 age-specific curves (at five year intervals between the ages of 25 and 95, with decreasing relative risk at increasing age) are used to characterize relative risk of IHD and strokes.

Relative risks obtained by applying the IERs to ground level  $PM_{2.5}$  concentrations from the GEOS-Chem results are used to determine the fraction of total incidence for each health endpoint per grid-cell attributable to emissions from each economic sector. This is done by first computing the PM2.5-attributable fraction of total incidence for all sectors using the GEOS-Chem runs with all sectors eliminated or reverted to 2010, and then apportioning the result by sector on the basis of change in PM2.5 attributable to each sector from the GEOS-Chem runs with individual sectors eliminated or reverted to 2010. This method ensures that the non-linearity of the Burnett IERs, which yield lower marginal relative risk with increasing PM<sub>2.5</sub> exposure, does not result in preferential treatment of any individual sector. This two-step process is summarized in equations **(2.5)** and **(2.6)**.

Total Sectoral Contribution (All Sectors, 2015):

$$
\Delta I_h(x, y) = I_h \left( 1 - \frac{RR_{h, \text{off}}(x, y)}{RR_{h, 2015}(x, y)} \right)
$$
\n(2.5)

Differential Sectoral Contribution (All Sectors, 2015-2010):

$$
\Delta I_h(x, y) = I_h \left( 1 - \frac{RR_{h,2010}(x, y)}{RR_{h,2015}(x, y)} \right)
$$
\n(2.6)

where:

 $I_h$  is the total incidence of health endpoint  $h$  $\Delta I_h$  is the PM<sub>2.5</sub>-attributable fraction of  $I_h$  due to all sectors  $RR_{h,off}$  is relative risk with all Asia-Pacific sectoral emissions eliminated  $RR<sub>h,2010</sub>$  is relative risk with all Asia-Pacific sectoral emissions at 2010 values  $RR<sub>h.2015</sub>$  is relative risk with all Asia-Pacific sectoral emissions at 2015 values  $x, y$  are (2-D) spatial indices of the ground-level GEOS-Chem PM<sub>2.5</sub> distribution

 $\Delta I_h$  is then apportioned by sector through equation **(2.7)** 

$$
\Delta I_{c,h}(x,y) = \Delta I_h(x,y) \frac{\Delta P M_{2.5,c}(x,y)}{\sum_{i=1}^{i} \Delta P M_{2.5,i}(x,y)}
$$
(2.7)

where:

 $\Delta l_{c,h}$  is the incidence of health endpoint h attributable to each sector c

 $i_{\text{sect}}$  is the number of sectors

 $\Delta PM_{2.5}$  is change in PM2.5 by eliminating or reverting emissions from individual sectors

and all other variables are consistent with equations (2.5/2.6)

For this study country-specific incidence rates of the five identified health endpoints  $(I_h)$  for 2010 and 2015 at all age brackets relevant for the WHO GBD 2016 IERs are obtained from the WHO GBD 2015 study [51]. These incidences are applied in Equations (5a/5b) to compute fractional incidences attributable to each sector, which are subsequently multiplied by the relevant population (ages 25+ for IHD and strokes, and all ages for COPD, LRI, and lung cancer) in each grid-cell to compute mortalities. The maps of population distribution used in the study are sourced from the LandScan 2010 and 2015 datasets provided at 30 arc-second resolution globally by Oak Ridge National Laboratory [37, 38].

Premature mortalities are computed both regionally and globally. Regional premature mortalities are computed for the 10 most populous countries in the Asia-Pacific region as defined in **Figure 2-1**: Bangladesh, China, India, Indonesia, Japan, Myanmar, Philippines, South Korea, Thailand, and Vietnam (together, constituting more than 95% of the Asia-Pacific population), as well as for the remainder of the region combined. Global impacts are quantified both in total for the whole world, and also on a sub-regional basis for 8 different regions including Africa, Asia-Pacific, Commonwealth of Independent States (CIS), Europe, Middle East, North America, Oceania, and South America. While Eurasia is split into four regions (Asia-Pacific, CIS, Europe, and Middle East) in order to distinguish between near-field and far-field impacts stemming from the Asia-Pacific, the remaining regions of Africa, North America, Oceania, and South America are defined according to continental grouping. A map further clarifying the definition and borders of each of these global regions is presented in **Figure A-2**.

### **3 Results**

We first discuss the evolution of sectoral emissions from the Asia-Pacific region between 2010 and 2015. This includes identification of the major activity and policy drivers of the emissions trends (section **3.1**) and comparison of the simulated changes in aerosol optical depth (AOD) against satellite measurements (section **3.2**). Subsequently, the effect of these recent changes in Asia-Pacific sectoral emissions in terms of contribution to both total global and regional mortalities in 2015 as well as contribution to change in such mortalities between 2010 and 2015 are quantified (section **3.3**). Using these results, the effectiveness of individual abatement measures modeled as part of the updated inventory in terms of reducing PM<sub>2.5</sub>-attributable mortalities is evaluated and sectors and countries in this region causing or experiencing highest growth in  $PM<sub>2.5</sub>$ -attributable mortalities are identified as the most pressing targets for future mitigation strategies to address.

### **3.1 Emissions trends**

Changes in ground-level distribution and population exposure to  $PM_{2.5}$  are principally driven by changes in emissions of total primary PM2.5 (consisting of BC, OC, primary sulfates, nitrates, ammonium, and trace metals) and the three major chemical precursors of secondary PM<sub>2.5</sub> (NH<sub>3</sub>,  $NO<sub>x</sub>$ , and  $SO<sub>2</sub>$ ), as these are the major sources of PM<sub>2.5</sub> into the atmosphere. Annual emissions of each of these chemical species between 2010 and 2015 are shown in **Table 3.1** for the three major emitter countries of China, India, and Indonesia (representing 80-83% of total

anthropogenic NH<sub>3</sub>, NO<sub>x</sub>, primary PM<sub>2.5</sub>, and SO<sub>2</sub> emissions in the region), as well as for the remainder of the region combined.



**Table 3.1:** Evolution of emissions of primary PM2.5 and PM2.5 precursors between 2010 and 2015 from China, India, Indonesia, and other Asia-Pacific countries in Tg (species)/year

The emissions trends vary by species and location. These trends are attributable to both changes in human activity and fuel use by sector, as well as government-driven measures to introduce emissions controls through both legislative and technological means. This is visible in Figure 3-1, which shows spatial variations in the changes in NH<sub>3</sub> emissions from the agricultural sector and  $NO<sub>x</sub>$  emissions from three emitting sectors (energy, railroads, and road transportation) with unique spatial patterns of change.

The broad increases in ammonia emissions visible in **Figure 3-1(a)** are driven by growth in the agricultural sector, specifically of fertilizer use and livestock production. Growth is particularly

pronounced in South and Southeast Asia, especially in the major agricultural producers Bangladesh, Indonesia, and Vietnam (34%, 22%, and 31% growth respectively). Conversely, agricultural ammonia emissions from Japan and Malaysia are found to fall over this period, undergoing reductions of 9% and 7% respectively between 2010 and 2015. These reductions in both countries are purely driven by changes in fertilizer use between 2010 and 2015, as reported by the UN FAO [14]. Growth in NH<sup>3</sup> emissions from India also exhibit a different pattern of growth from other countries, with a 9% decrease in annual emissions modeled between 2011 and 2013, also driven by a dip in fertilizer use between these years. This disruption in the periods of growth observed for 2010-2011 and 2013-2015 may be attributable to changes in Indian government policies on agricultural subsidies, including an 11.3% cut in subsidies for nitrogen-based fertilizers between 2012 and 2013 [34], which may have affected fertilizer usage in the short-term.

Unlike ammonia, changes in emissions of  $NO<sub>x</sub>$ , primary  $PM<sub>2.5</sub>$ , and  $SO<sub>2</sub>$  show a polarized pattern across the Asia-Pacific region, as shown in **Figure 3-1(b)**. NO<sub>x</sub> emissions for India and Indonesia increase by 29% and 27% respectively, primarily driven by increased activity in the energy, manufacturing, and residential sectors, with no new emission controls applied for these sectors in these two countries over the time period of interest. This represents a total increase in NO<sub>x</sub> emissions of 3.0 Tg/year from these two countries between 2010 and 2015, which is less than the reduction in  $NO_x$  emissions from China of 3.7 Tg/year over the same period, but also more than 36% of total  $NO<sub>x</sub>$  emissions in 2015 from all other Asia-Pacific countries. The reduction in Chinese  $NO<sub>x</sub>$  emissions represents a decrease of 15% relative to 2010. This decrease is almost entirely driven by the introduction of selective catalytic reduction (SCR) technologies into Chinese coal-fired power plants, resulting in a 60% reduction of corresponding emission factors according to Liu et al. [32]. Despite an increase of 17% in use of coal by the Chinese energy sector over these five years, the introduction of SCR results in a net reduction of 40% of  $NO<sub>x</sub>$  emissions from this sector, and results in emissions of  $NO<sub>x</sub>$  from China peaking in 2011. This trend in Chinese NO<sub>x</sub> emissions is consistent with existing literature by Liu et al. [30], which used satellite observations to determine a peak in column-total  $NO<sub>x</sub>$  concentrations over China in 2011.



Figure 3-1. Changes in Asia-Pacific emissions of agricultural NH<sub>3</sub> in units of Mg(N)/km<sup>2</sup>/year, and NO<sub>x</sub> from the energy, railroad, and road transportation sectors in units of Mg(NO<sub>2</sub>)/km<sup>2</sup>/year between 2010 and 2015.

 $NO<sub>x</sub>$  emissions from the rest of the Asia-Pacific region are also shown to undergo minor reductions of 2-3% or -0.2 Tg/year in total over the entire period between 2010 and 2015. This is especially noticeable in South Korea, where annual  $NO<sub>x</sub>$  emissions from road vehicles fell by 0.5 Tg/year between 2010 and 2015 due to the implementation of more stringent vehicle emission standards. As seen in **Figure 3-1(d)**, this policy-driven effect on the road transportation sector is not limited to South Korea, but present across most of the region with the exception of some countries in Southeast Asia. As road vehicle emissions of  $NO<sub>x</sub>$  comprise more than 16% of total regional  $NO<sub>x</sub>$  emissions in 2010, the effect of region-wide implementation of advanced vehicle emissions standards constitutes a second key driver for these emissions.

Emissions of both total primary  $PM_{2.5}$  and  $SO_2$  also follow the polarized pattern observed in **Figure 3-1(b)**, with reductions in China and a net increase in the remainder of the region. Reductions of these species in China are driven by the energy, manufacturing, and residential sectors, and attributable to several mitigative measures adopted as part of China's Action Plan on the Prevention and Control of Air Pollution in effect since 2013 [9, 58]. Emissions of PM<sub>2.5</sub> and SO<sub>2</sub> from the Chinese energy sector are estimated to fall by 25% and 50% respectively due to a range of measures including improved operationalization of flue gas desulfurization in power plants and promoted use of higher quality coal with lower ash and sulfur content [58]. In the case of the Chinese manufacturing sector, other measures such as phase out of small coal-fired industrial boilers in urban areas, desulfurization of emissions from large industrial boilers through sorbent injection, installation of electrostatic precipitators to remove primary PM<sub>2.5</sub> emissions from manufacturing plants, and replacement of old manufacturing capacity with new plants of higher combustion efficiency, result in reductions of  $PM<sub>2.5</sub>$  and  $SO<sub>2</sub>$  emissions by 28% and 41% respectively [58]. Reductions of 23% in primary PM<sub>2.5</sub> emissions and 19% in SO<sub>2</sub> emissions from the Chinese residential sector are driven by introduction of improved technology such as cleanerburning stoves [58]. Furthermore, shifts in fuel use towards cleaner burning forms of coal, such as briquettes, also aid emissions reduction in both these sectors in China [58]. Combined, mitigative measures applied to these three sectors result in reductions of 17% (-2.2 Tg/year) and 35% (-10.5 Tg/year) of primary  $PM<sub>2.5</sub>$  and  $SO<sub>2</sub>$  emissions from China respectively. These

reductions exceed growth in emissions of these two species in the rest of the region, despite high net growth (21% and 47% respectively) in India.

Apart from these major sectoral drivers of emissions changes, many other drivers (both explicitly and implicitly modeled) from other individual sectors contribute to the total change in emissions over the five year period of interest. The effect of more stringent road vehicle emissions standards is not limited to reduction of just  $NO<sub>x</sub>$  emissions, but also of emissions of CO, NMVOCs, and primary PM2.5. Improved fuel quality standards implemented for the road transportation sector across this region over this period similarly reduce  $SO<sub>2</sub>$  emissions from this sector. Furthermore, changes in diesel fuel use by the rail transportation sector lead to different trends in emissions of all species by this sector across the region, as visualized in **Figure 3-1(c)**. While strong growth in diesel use by railways leads to growth in South Asian rail emissions, specifically by 15% in India and 70% in Bangladesh, declining diesel use in other parts of the region such as East and Southeast Asia, attributed to electrification of railways and phase-out of operating diesel-fired rolling stock, leads to emission reductions from the rail sector of 41%, 7%, and 39% in China, Japan, and South Korea respectively.

### **3.2 Validation**

We compare simulated aerosol optical depth (AOD) from GEOS-Chem to satellite observations from NASA's Moderate Resolution Imaging Spectroradiometer (MODIS) instruments [36]. For these comparison runs, regional wildfire emissions were updated to 2015 values to yield a better match against the observed spatial distribution of AOD. This is not the case for the sectoral impact evaluation runs used to compute premature mortalities, as this study does not examine changes in PM2.5-driven mortalities due to non-anthropogenic sources such as wildfires. **Figure 3-2** shows the simulated and observed annual average AOD across the region in 2015, as well as the change in AOD between 2010 and 2015.

GEOS-Chem runs successfully capture the broad spatial pattern and magnitude of aerosols over the Asia-Pacific region in 2015, including particularly high concentrations over the Chinese industrial heartland in eastern China and the Indo-Gangetic plain in northern India. Individual features, such as the distinct hotspot of aerosol formation over the Chongqing-Chengdu urban



#### **(a) Asia-Pacific AOD in 2015**

**Figure 3-2:** Comparison of AOD results from MODIS satellite observational data and GEOS-Chem simulations for 2015 and change between 2010 and 2015.

 $0.2$ 

 $-0.2$ 

 $-0.1$ 

 $\mathsf{O}\xspace$ 

 $\triangle AOD$  (-)

 $0.1$ 

 $0.2$ 

 $0$ <br> $\triangle AOD$  (-)

 $-0.2$ 

 $-0.1$ 

 $0.1$ 

areas in South-Central China are also captured in the GEOS-Chem runs. Comparison of the change in AOD between the two years shows that the GEOS-Chem runs capture the broad trends of increasing AOD in India, decreasing AOD in China, increases over the majority of the Southeast Asia sub-region, and minor decreases in remaining East Asian countries such as South Korea and Japan.

Disagreement between AOD in 2015 according to GEOS-Chem and MODIS results is particularly prominent in western China and parts of Southeast Asia including Indonesia, Malaysia, Brunei, and the Philippines. The disagreement in these parts of the Asia Pacific are driven by non-anthropogenic sources that lie outside the scope of this study and therefore do not affect final results. These include (respectively) changes not modeled in geogenic dust emissions from the Gobi and Taklimakan deserts in north-central and western China and limited spatial accuracy of the GFED4 wildfire inventory used to model wildfires in 2015 across Southeast Asia. Furthermore, in regions of low aerosol concentration, MODIS observational data becomes less reliable as noise becomes a large fraction of the measurement signal.

Although these disagreements between GEOS-Chem and MODIS can cause high local differences in AOD, our focus is on the change in population exposure. As a proxy for the error in population exposure, we estimate the error in population-weighted AOD across the entire Asia-Pacific region. We find that for 2015, the error in GEOS-Chem values of population-weighted AOD relative to MODIS values is 11%, well within typical error bounds for modeling studies in this region [17, 21]. Furthermore, for the five most populous countries in this region of Bangladesh, China, India, Indonesia, and Japan, which account for more than 85% of the regional population, error ranges from -6% to 33%. In the two most populous countries of China and India, which contain over 70% of the regional population, the mean error is -6% and 16% respectively. For the other three countries, error is 13% for Japan, and 33% for both Bangladesh and Indonesia. **Table A.1** lists the population-weighted AOD both region-wide and from each of these country groups as obtained from GEOS-Chem and MODIS.

The comparison of change in regional AOD between 2010 and 2015 as per GEOS-Chem and MODIS also exhibits several differences, particularly across Southeast Asia, where GEOS-Chem underestimates AOD over areas due to limitations in modeling 2015 wildfires, and also along the

Asia Pacific-Russia border stretching from Mongolia to northern Japan. The latter is attributed to non-regional sources, including changes in both anthropogenic emissions and wildfire activity in Russia, which are also outside the scope of this study. For change in population-weighted AOD between 2010 and 2015, the region-wide error is 27%. Error over Bangladesh, China, and India are found to be lower than the region-wide value, at -6%, -10%, and 13%, while higher errors of 50% and 60% are observed over Indonesia and Japan respectively. **Table A.2** lists the change in population-weighted AOD both region-wide and from each of these countries as obtained from GEOS-Chem and MODIS.

The elevated error in Indonesia is attributed to the limitations in wildfire modeling, and that in Japan is attributed to the relative low change in AOD it experiences between 2010 and 2015, which renders the calculation of change in AOD using the MODIS dataset inaccurate due to noise interference. However, comparison of the change in ground-level  $PM_{2.5}$  concentration for the Tokyo metropolitan area between 2010 and 2015 yields values of -0.6  $\mu$ g/m<sup>3</sup> respectively from GEOS-Chem results, and -0.9  $\mu$ g/m<sup>3</sup> respectively from ground-level observational data [18, 43]. This suggests that GEOS-Chem is able to capture change in aerosol distribution over the most populated regions of Japan, both in terms of sign and magnitude with an error on the order of 33%.

### **3.3 Mortality impacts**

The changes in emissions from the Asia-Pacific region between 2010 and 2015 lead to changes in human exposure to ambient  $PM_{2.5}$  and resulting premature mortalities. Each sector's contribution to the change in both global and regional mortalities between 2010 and 2015 is presented in this section. The sectoral contribution to total mortalities is also quantified to contextualize the effect of the changes between 2010 and 2015 and identify shifts in the relative impact of individual sectors. Furthermore, the effect of abatement measures on changes in the mortality burden are assessed and sectoral targets for future mitigation strategies are identified.

Globally, a total of 1,750,000 (95% CI: 1,520,000-1,970,000) and 2,000,000 (95% CI: 1,740,000-2,260,000) PM<sub>2.5</sub>-driven premature mortalities are attributed annually to anthropogenic emissions stemming from the Asia-Pacific region in 2010 and 2015 respectively. 98% of these mortalities (1,710,000 and 1,960,000 mortalities respectively) are found to occur in the Asia-Pacific region itself, and the remaining 2% are exported to other regions. The total mortalities computed for these 2 years yield a change of 250,000 annual global premature mortalities due to Asia-Pacific anthropogenic emissions between 2010 and 2015. However, only 14% of this total change, or 34,000 (95% CI: 23,000-47,000) additional annual global premature mortalities, are attributable to change in ambient concentrations of  $PM_{2.5}$ , with the remaining 86% attributable to changes in demographics and socioeconomics, including population growth, migration, aging, and change in total incidence of the health endpoints linked to  $PM_{2.5}$  exposure. **Figure A-3** summarizes the breakdown of total mortalities due to each of these drivers. As drivers other than  $PM_{2.5}$  concentration are outside the scope of regulation by emissions controls, we only report change in mortalities due to change in PM<sub>2.5</sub> exposure.

#### **3.3.1 Regional impacts**

Of the 1,960,000 annual  $PM_{2.5}$ -driven mortalities stemming from Asia-Pacific anthropogenic emissions and occurring within the Asia-Pacific region in 2015, the vast majority are found to occur in China and India, with 904,000 (95% CI: 788,000-1,020,000) and 702,000 (95% CI: 608,000-798,000) mortalities respectively. Together, these two countries, which contain around 70% of the regional population, account for more than 80% of the regional burden of  $PM_{2.5}$ driven mortalities in 2015. These results are also of the same order and within the range of  $PM_{2.5^-}$ driven mortalities previously calculated for these countries, including by Lelieveld et al. [26] and by Cohen et al. [10], as presented in **Table A.3**. Smaller burdens of 67,000 (95% CI: 58,000-75,000); 92,000 (95% CI: 82,000-102,000); 44,000 (95% CI: 34,000-56,000); 25,000 (95% CI: 21,000-29,000); and 31,000 (95% CI: 28,000-35,000) mortalities per year are also found to occur in Bangladesh, Indonesia, Japan, Thailand, and Vietnam respectively, which also rank among the 20 most populous countries in the world. The remainder of the regional burden is dominated by other relatively populous countries such as Myanmar, the Philippines, and South Korea.

Figure 3-3(a) illustrates both the total PM<sub>2.5</sub>-driven mortalities per year in each of these receptor countries, as well as the sectoral breakdown in each. Emissions from the agricultural



**Figure 3-3:** Sectoral contributions to total annual PM2.5-driven mortalities in Asia-Pacific receptor countries in 2015 and change between 2010 and 2015 in 1,000s of mortalities/year.

sector, including agricultural waste burning, are the leading source of  $PM_{2.5}$ -driven premature mortalities, both at a regional and national level for most of the receptor countries with the exception of Vietnam, where residential emissions have a roughly equal effect. Ambient  $PM_{2.5}$ from agricultural emissions are found to cause around  $606,000$  (95% CI: 527,000-684,000) mortalities per year across the region, more than 30% of the total regional burden. This is followed by the industrial (including manufacturing and industrial process emissions), residential, and energy sectors, accounting for 436,000 (95% CI: 328,000-490,000), 420,000 (95% CI: 365,000-475,000), and 320,000 (95% CI: 279,000-362,000) mortalities respectively. Among the remaining sectors with lower impacts, road transportation and fuel processing emissions also contribute significant impacts of 73,000 (95% CI: 64,000-83,000) and 58,000 (95% CI: 51,000-66,000) annual mortalities respectively.

Regional growth in PM<sub>2.5</sub>-driven mortalities between 2010 and 2015 is driven by opposing changes in emissions from individual countries, whose major drivers and trends differ from those of total regional impacts in 2015, as shown in **Figure 3-3(b)**. Across South and Southeast Asia, sustained economic and activity growth lead to  $125,000$  (95% CI:  $103,000-163,000$ ) additional annual premature mortalities regionally in 2015 compared to 2010, primarily impacting India, Indonesia, and Bangladesh. However, other sectors and parts of the region undergo emissions reductions due to falling activity rates from structural changes such as electrification of railroads, and emissions controls from regulations such as China's Action Plan on the Prevention and Control of Air Pollution [9], which targeted the Chinese energy, industrial, and residential sectors, as well as region-wide adoption of Euro IV/V/VI-compliant road vehicle emission and fuel quality standards. These abatement measures lead to a total reduction of 94,000 (95% CI: 76,000-128,000) annual premature mortalities, primarily across East Asia, including China, Japan, and South Korea. These opposing trends lead to a net increase in regional PM<sub>2.5</sub>-driven mortalities of 31,000 (95% CI: 21,000-43,000), or 1.7%, due to changes in ambient PM<sub>2.5</sub> concentrations between 2010 and 2015.

While total 2015 impacts in China exceed those in India, computed change in impacts between 2010 and 2015 show mortalities in China to have decreased by 70,000 (95% CI: 60,00094,000), or 7%, over the five-year period, in contrast to a net increase of 83,000 (95% CI:

67,000-110,000) annual mortalities in India (13%). The magnitude of reductions in China alone exceeds the net growth in mortalities in all other countries excluding India combined, including 7,000 (95% CI: 6,000-8,000) and 11,000 (95% CI: 10,000-13,000) additional annual mortalities in Bangladesh and Indonesia respectively. The high growth in mortalities in India stems from the energy and industrial sectors in particular, which are found to cause an additional 30,000 (95% CI: 24,000-40,000) and 41,000 (95% CI: 34,000-50,000) annual mortalities respectively, or over 85% of the net national growth. The growth in impacts affecting India from the industrial sector (which exhibits high collocation with population centers), in particular, is the highest across the entire region for a single sector in a single receptor country. The high contribution of these sectors in India to both total 2015 impacts and growth in impacts between 2010 and 2015 make them leading sources to target with emissions controls as part of future abatement measures. Other sectors, such as the agriculture and residential sectors, which have a greater contribution to total mortalities in 2015, contribute less to changes between 2010 and 2015, causing 22,000 (95% CI: 18,000-27,000) more and 15,000 (95% CI: 13,000-20,000) fewer annual mortalities region-wide respectively.

The reduction in PM2.5-driven premature mortalities in China observed between 2010 and 2015 is likely due to the emissions controls imposed on the energy, industrial, and residential sectors in this five-year period. In the Chinese energy sector, the introduction of SCR, transition to cleaner variants of coal with lower ash and sulfur content, and expansion of flue gas desulfurization in Chinese power plants [58] between 2010 and 2015 to reduce  $NO<sub>x</sub>$ , primary  $PM_{2.5}$ , and SO<sub>2</sub> emissions contributes to 34,000 (95 % CI: 29,000–45,000) fewer mortalities in 2015 than 2010 in China alone. As energy generation capacity grew close to 38% between 2010 and 2015, from 4,200 TWh to 5,800 TWh [35], this means that mortalities per unit of energy generated fell by 47%, from 34 mortalities/TWh in 2010 to 18 mortalities/TWh in 2015 if only direct emissions from power plants are considered.

For the Chinese industrial sector, the phase-out of small coal-fired industrial boilers and outdated industrial capacity as well as desulfurization and particulate filtering in other industrial facilities contributes to a slightly lower reduction of 25,000 (95% CI: 18,000-37,000) fewer annual mortalities in 2015 compared to 2010. Lastly, introduction of technologies such as clean-burning

stoves as well as cleaner variants of fuels in the Chinese residential sector contributes to 21,000  $(95\%$  CI: 18,000-28,000) fewer annual mortalities in 2015 than 2010. The reductions in mortalities achieved in any of these three sectors alone completely offsets the growth in mortalities in China from all other sectors. Furthermore, the reduction in mortalities from the Chinese energy and residential sectors also exceed the rise in mortalities from the corresponding sector in India. However, this is not the case for the industrial sector, where growth in India outstrips reductions in China by 16,000 annual mortalities over this five-year period. These results illustrate the high region-wide mitigative impact attainable through controls imposed on individual PM2.5 precursor species from the energy, industrial, and residential sectors, further suggesting the benefits of such mitigation applied to the Indian energy sector as part of future regulation.

Spatial distributions of the change in  $PM<sub>2.5</sub>$  across East Asia from the energy and industrial sectors show that the imposition of emission controls for these sectors in China has contributed to transboundary benefits. We estimate reductions in mortalities attributable to these sectors in neighboring countries such as South Korea (12% fewer mortalities per year due to energy emissions) despite growth in local energy sector emissions (6-8% increase in energy sector emissions from South Korea). Transboundary benefits are likely to be even more pronounced in the case of application of such measures in India owing to the proximity of highly populous neighboring countries both within the Asia-Pacific region and in neighboring regions, such as Bangladesh and Pakistan respectively.

Regional impacts from the transportation industry are also found to have fallen over this time period. Reductions in emission from the rail and road transportation sectors result in 1,400 (95% CI: 1,200-1,800) and 4,900 (95% CI: 4,000-6,400) fewer mortalities in 2015 compared to 2010 across all countries in the region. The trend in mortalities by country due to rail transportation mirrors the trend observed for the energy sector, with reductions in annual mortalities observed for China, and increases observed in India. Although no abatement measures are explicitly modeled for the rail transportation sector, the reduction of sector-attributable mortalities in China by  $1,900$  (95% CI:  $1,600-2,600$ ) mortalities/year could be driven by increasing electrification or modernization of the Chinese railway locomotive fleet. The region-wide

reduction in mortality due to rail and road transportation emissions is partially counteracted by growth in diesel fuel use by the Indian rail sector, with 500 (95% CI: 400-600) additional premature mortalities annually in India from these sectors. Furthermore, while measures such as electrification transfer combustion from the rail sector to the energy sector, in the case of countries such as China, this transfer is not manifested as a transfer of emissions due to the effects of new emissions controls and other improvements applied to the local energy sector.

Changes in PM2.5-driven premature mortalities from the road transportation sector are more uniform across the region, as most countries, with the exception of Indonesia and a few other Southeast Asian countries, implemented more stringent vehicle emission and fuel quality standards for both gasoline and diesel fuels. This ranged from Euro II/III-equivalent standards in some countries (e.g. Indonesia and India respectively) in 2010, to Euro IV/V/VI-equivalent standards in most countries in 2015 (excepting previously identified countries in Southeast Asia, which maintain Euro II/III-equivalent standards throughout). The improvements in emission and fuel quality standards not only offset all growth in emissions that might have occurred as a result of activity growth in this sector over the five-year period, but also result in up to 3,200 (95% CI:  $2,700-4,200$ ) and  $1,200$  (95% CI:  $1,000-1,600$ ) avoided deaths in China and India respectively.

#### **3.3.2 Global impacts**

The spatial distributions of annual mortalities due to total Asia-Pacific anthropogenic emissions in 2015 and changes thereof between 2010 and 2015 are presented in **Figure 3-4** on a per unit area basis. The results for the Asia-Pacific region are obtained from fine resolution GEOS-Chem runs at  $0.5^{\circ} \times 0.666^{\circ}$  and are overlaid on results for the rest of the world obtained from the coarse  $4^{\circ} \times 5^{\circ}$  resolution runs. Results obtained for the Asia-Pacific region from the coarse resolution runs are found to agree within 20% of results obtained from fine resolution runs, with the grid-dependent error decreasing with distance from the region as a result of dispersion of transported emissions and impacts. This error margin lies within the 95% confidence interval of results in this study.



#### **(a) Premature mortalities / sq. km due to total Asia-Pacific 2015 sectoral emissions**

**(b) Premature mortalities / sq. km due to change in Asia-Pacific sectoral emissions (2010-15)**



Figure 3-4: Annual global PM<sub>2.5</sub>-driven premature mortalities due to Asia-Pacific anthropogenic emissions in 2015 and change in emissions between 2010 and 2015, in units of mortalities/km<sup>2</sup>/year.

Results for the rest of the world show export of mortality impacts due to Asia-Pacific emissions to four of the six other global regions, with the exception of Oceania and South America. Of the 38,000 (95% CI: 33,000-43,000) exported premature mortalities due to exposure to PM2.5 attributable to Asia-Pacific anthropogenic emissions in 2015 (2% of total impacts), over 28,000, or 74%, are estimated to occur in the Middle East, predominantly in highly populated countries bordering the Asia-Pacific, such as Pakistan. Lower burdens of 5,300 (95% CI: 4,500-6,200), 3,500 (95% CI: 3,000-4,000), 500 (95% CI: 400-600), and 400 (95% CI: 300-500) mortalities are estimated for the CIS, Europe, North America, and Africa regions respectively. The export of PM2.5-driven premature mortalities from the Asia-Pacific region is also found to increase by 2,800 (95% CI: 1,800-3,900) between 2010 and 2015, with 3,000 additional premature mortalities exported to the Middle East in 2015 compared to 2010, between 100-200 fewer mortalities exported to CIS and North America each, and up to 100 additional mortalities exported to Africa and Europe each.

In terms of sectoral contribution to global impacts, the exported mortalities are attributed to emissions from agriculture, with 11,000 (95% CI: 9,600-12,000) annual exported mortalities; energy, with 9,200 (95% CI: 8,100-10,000) exported mortalities; the residential sector, with 7,100 (95% CI: 6,200-8,000) annual exported mortalities; industry, with 6,300 (95% CI: 5,400-7,100) annual exported mortalities; and road transportation, with 2,600 (95% CI: 2,200-2,900) annual exported mortalities. Combined, these sectors account for over 95% of the exported mortalities. The shares of exported mortalities attributed to each sector differ from the shares of Asia-Pacific mortalities. While the industrial sector exceeds the residential sector and the latter exceeds the energy sector as sources of mortalities caused within the Asia-Pacific region, this ranking is reversed for exported impacts, with the energy sector causing 2,100 and 2,900 more mortalities outside the region than the residential and industrial sectors respectively.

This reversal can be attributed to two characteristics of the energy sector, namely, the higher proportion of secondary PM2.5 precursors emitted and spatial distribution near border areas of the Asia-Pacific region. The higher proportion of  $PM_{2.5}$  precursor emissions compared to primary PM<sub>2.5</sub> means that this sector has greater impact through long-range pathways of PM<sub>2.5</sub> formation, making its impacts more amenable to export compared to the residential and industrial sectors,

whose impacts are dominated by primary  $PM<sub>2.5</sub>$  and thus more local. Furthermore, energy emission sources are more prevalent than manufacturing or residential sources in both the western fringes of India that border Pakistan as well as in the northern region of China bordering Russia, also making it easier for  $PM_{2.5}$  impacts from this sector to spill into neighboring regions.

Sectoral contribution to growth in exported mortalities between 2010 and 2015 also differs from contribution to total exported mortalities in 2015. While the agriculture sector is the leading source of total exported mortalities in 2015, growth in Asia-Pacific energy and industrial emissions acts as the primary driver of growth in exported mortalities, resulting in 1,200 (95% CI: 800-1,700) and 1,100 (95% CI: 700-1,500) more deaths exported in 2015 than 2010 respectively. The growth of these two sectors in South Asia (mostly in India) in particular, is the primary driver of the increase in exported mortalities, as most of the additional impacts occur in the neighboring Middle East region. While the residential sector is the third largest contributor to total exported mortalities in 2015, it does not contribute at all to change between 2010 and 2015. This is also driven by the trend of emissions in South Asia, where the residential sector show little change between 2010 and 2015.

Emission controls applied across the Asia-Pacific region over this time period also affect exported mortalities. The effect of the controls applied to the energy, industrial, and residential sectors in China is discernable in changes to mortalities exported to the neighboring CIS region, where the energy sector causes 200 fewer annual exported mortalities in the CIS region in 2015 compared to 2010, and the industrial and residential sectors cause 100 fewer mortalities each. In addition, improved vehicle emission and fuel quality standards imposed on Asia-Pacific road transportation result in 100 fewer exported mortalities from this sector in 2015 compared to 2010, exceeding the entire growth in exported mortalities from the residential sector.

### **4 Conclusions**

Between 2010 and 2015, emissions of primary  $PM_{2.5}$  and  $PM_{2.5}$  precursors from the Asia-Pacific region show a polarized pattern of change driven by growth of up to 40-60% in South and Southeast Asian countries such as India and Indonesia, and reductions of up to 30-40% in several East Asian countries, including China and Japan due to activity pattern and regulatory changes including emission controls enacted as part of China's 2013 Action Plan on the Prevention and Control of Air Pollution. Regional emission reductions in rail and road transportation sectors are also achieved through both structural changes such as railroad electrification, and regulatory changes such as introduction of more stringent road vehicle emission and fuel quality standards. While global PM<sub>2.5</sub>-driven premature mortalities arising due to sectoral emissions from this region grow from 1,750,000 annually in 2010 to 2,000,000 annually in 2015, the emission controls applied in this region over this period help restrict the net ambient  $PM_{2.5}$  concentration-driven growth to 34,000 additional annual global mortalities between these two years. Where applied, these controls contribute to a total of 95,000 avoided annual premature mortalities in 2010 compared to 2015, partially offsetting 129,000 additional mortalities generated due to growth in emissions elsewhere in the region. While the former benefits are found to occur primarily across East Asia in countries such as China, Japan, and South Korea, the latter damages particularly impact highly populous countries across South Asia such as Bangladesh, India, and Indonesia. This pattern of changes in impacts is consistent with the understanding of changes in emissions in this region.

In both years, 98% of these mortalities occur within the Asia-Pacific region while the remaining 2% are exported to the rest of the world, particularly to neighboring regions such as the Middle East and the CIS countries. Spatially, China and India act as receptors for 80% of the regional impacts, with other populous countries such as Bangladesh, Indonesia, Japan, Thailand, and Vietnam also incurring between 25,000 and 100,000 annual mortalities each. Sectorally, the agricultural, industrial, and residential sectors are found to cause the most region-wide mortalities in 2015, accounting for 30%, 22%, and 21% of the regional total. The energy and industrial sectors are the leading contributors to gross increase in regional premature mortalities between 2010 and 2015, contributing a combined 71,000 additional mortalities in India alone, or over 55% of the total of 129,000 additional mortalities. Alongside the agricultural sector, the energy sector is also a dominant contributor to mortalities exported from the Asia-Pacific region to the remainder of the world, and serves as the leading contributor to growth in exported mortalities between 2010 and 2015.

This study leaves scope for future work in several areas. As a scaling approach is deployed to obtain updated 2015 emissions for the Asia-Pacific region in this study, the effect of spatial displacement as a possible abatement measure, through shifting of emission sources away from population centers, between 2010 and 2015 is not analyzed here. Analysis of such measures requires development of updated spatial surrogates for this region specific to year-2015 as a prerequisite, and it is hoped that such information will become available for future work in quantifying the effectiveness of such measures.

The net increase of 83,000 additional annual  $PM_{2.5}$ -driven premature mortalities in India between 2010 and 2015, in contrast to the reduction in China of 70,000 fewer annual premature mortalities confirms that in the present decade, India has supplanted China as the primary receptor of growth in health impacts due to ambient  $PM_{2.5}$  air pollution in this region. Therefore, while the characterization of air quality impacts in China has constituted the bulk of research on understanding the health impacts of air pollution in Asia in the past, the results of this study show that an equally detailed characterization of air quality impacts in India is crucial in informing future mitigation policies for this region.

The counteracting trends of  $PM_{2.5}$ -driven premature mortalities in China and India also show that quantification of air quality impacts purely at a regional level for this part of the world can be misleading, as the relatively low net concentration-driven growth for Asia-Pacific as a whole

between 2010 and 2015 obscures much higher magnitudes of growth and decline in individual countries. This suggests the need for greater understanding of shifts in air quality burden within the region, especially of transboundary transport and the source-receptor relationships of air pollutants in this region. Such a study would also help inform policymaking through evaluation of the transboundary effect of policy choices made by individual countries on other regional stakeholders. Furthermore, by assessing the relative burden of domestic and transboundary air pollution-driven health impacts, each individual receptor countries' degree of control over the primary sources of impacts within its borders could be determined. This would characterize the effectiveness of country-level mitigation strategies and inform the requirements for broader, region-wide mitigation strategies.

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## **Appendix**



**Figure A-1:** WHO GBD 2016 IERs for COPD, IHD, LRI, lung cancer, and strokes (mean & 95% confidence intervals) (Gakidou et al., 2017).



**Figure A-2:** Map of global regions as defined for this study.

**Table A.1:** Year-2015 population-weighted AOD in the Asia-Pacific region, as obtained from GEOS-Chem and NASA MODIS satellite observations



**Table A.2:** Change in population-weighted AOD in the Asia-Pacific region between 2010 and 2015, as obtained from GEOS-Chem and NASA MODIS satellite observations





Figure A-3: Total change in PM<sub>2.5</sub>-driven premature mortalities between 2010 and 2015 due to all drivers in Asia-Pacific receptor countries.

<b>Emission Year</b>	2000	2010	2013	2015
<b>Source</b>	Silva et al. (2013)	Lelieveld et al. (2015)	Hu et al. (2017)	Cohen et al. (2017)
<b>China</b>	1,252,000 (incl. other East Asia)	1,360,000	1,300,000	1,108,000
India	515,000	650,000	-	1,090,000

**Table A.3:** Existing estimates of annual premature mortalities due to exposure to outdoor air pollution in China and India

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# **Bibliography**

- [1] Alexander, B., 2005. Sulfate formation in sea-salt aerosols: Constraints from oxygen isotopes. Journal of Geophysical Research 110.<https://doi.org/10.1029/2004JD005659>
- [2] Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., Winiwarter, W., 2011. Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications. Environmental Modeling & Software 26, 1489–1501.<https://doi.org/10.1016/j.envsoft.2011.07.012>
- [3] Apte, J.S., Marshall, J.D., Cohen, A.J., Brauer, M., 2015. Addressing Global Mortality from Ambient PM2.5. Environmental Science & Technology 49, 8057–8066. <https://doi.org/10.1021/acs.est.5b01236>
- [4] BASF, 2017. BASF Report 2017: Economic, environmental, and social performance. BASF SE. URL: [https://www.basf.com/documents/corp/en/about](https://www.basf.com/documents/corp/en/about-us/publications/reports/2018/BASF_Report_2017.pdf)[us/publications/reports/2018/BASF\\_Report\\_2017.pdf](https://www.basf.com/documents/corp/en/about-us/publications/reports/2018/BASF_Report_2017.pdf)
- [5] Bey, I., Jacob, D.J., Yantosca, R.M., Logan, J.A., Field, B.D., Fiore, A.M., Li, Q., Liu, H.Y., Mickley, L.J., Schultz, M.G., 2001. Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation. Journal of Geophysical Research: Atmospheres 106, 23073–23095.<https://doi.org/10.1029/2001JD000807>
- [6] Binkowski, F.S., Roselle, S.J., 2003. Models-3 Community Multiscale Air Quality (CMAQ) model aerosol component 1. Model description. Journal of Geophysical Research 108. <https://doi.org/10.1029/2001JD001409>
- [7] Burnett, R.T., Pope, C.A., III, Ezzati, M., Olives, C., Lim, S.S., Mehta, S., Shin, H.H., Singh, G., Hubbell, B., Brauer, M., Anderson, H.R., Smith, K.R., Balmes, J.R., Bruce, N.G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M.C., Gapstur, S.M., Diver, W.R., Cohen, A., 2014. An Integrated Risk Function for Estimating the Global Burden of Disease Attributable to Ambient Fine Particulate Matter Exposure. Environmental Health Perspectives. <https://doi.org/10.1289/ehp.1307049>
- [8] Cecil, D.J., 2015. LIS/OTD Gridded Lightning Climatology Data Collection Version 2.3.2015.<https://doi.org/10.5067/LIS/LIS-OTD/DATA311>
- [9] China State Council, 2013. Action Plan on Prevention and Control of Air Pollution. China State Council, Beijing, China. URL: [http://www.gov.cn/zwgk/2013-](http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm) [09/12/content\\_2486773.htm](http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm)
- [10] Cohen, A.J., Brauer, M., Burnett, R., Anderson, H.R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C.A., Shin, H.,

Straif, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C.J.L., Forouzanfar, M.H., 2017. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015. The Lancet 389, 1907–1918[. https://doi.org/10.1016/S0140-](https://doi.org/10.1016/S0140-6736(17)30505-6) [6736\(17\)30505-6](https://doi.org/10.1016/S0140-6736(17)30505-6)

- [11] Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., Van Dingenen, R., Granier, C., 2016. Forty years of improvements in European air quality: regional policy-industry interactions with global impacts. Atmospheric Chemistry and Physics 16, 3825–3841. [https://doi.org/10.5194/acp-16-](https://doi.org/10.5194/acp-16-3825-2016) [3825-2016](https://doi.org/10.5194/acp-16-3825-2016)
- [12] Damian, V., Sandu, A., Damian, M., Potra, F., Carmichael, G.R., 2002. The kinetic preprocessor KPP-a software environment for solving chemical kinetics. Computers & Chemical Engineering 26, 1567–1579. [https://doi.org/10.1016/S0098-1354\(02\)00128-X](https://doi.org/10.1016/S0098-1354(02)00128-X)
- [13] FAA, 2016. Aviation Environmental Design Tool. Federal Aviation Administration Office of Environment and Energy. URL:<https://aedt.faa.gov/>
- [14] FAO, 2018. Food and Agricultural Organization of the United Nations: Statistics. Food and Agricultural Organization of the United Nations. URL: <http://www.fao.org/statistics/en/>
- [15] Fu, X., Wang, S., Zhao, B., Xing, J., Cheng, Z., Liu, H., Hao, J., 2013. Emission inventory of primary pollutants and chemical speciation in 2010 for the Yangtze River Delta region, China. Atmospheric Environment 70, 39–50. <https://doi.org/10.1016/j.atmosenv.2012.12.034>
- [16] Gakidou, E. et al., 2017. Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 1990–2016: a systematic analysis for the Global Burden of Disease Study 2016. The Lancet 390, 1345–1422. [https://doi.org/10.1016/S0140-6736\(17\)32366-8](https://doi.org/10.1016/S0140-6736(17)32366-8)
- [17] Geng, G., Zhang, Q., Tong, D., Li, M., Zheng, Y., Wang, S., He, K., 2017. Chemical composition of ambient  $PM_{2.5}$ ; over China and relationship to precursor emissions during 2005–2012. Atmospheric Chemistry and Physics 17, 9187–9203. <https://doi.org/10.5194/acp-17-9187-2017>
- [18] Hara, K., Homma, J., Tamura, K., Inoue, M., Karita, K., Yano, E., 2013. Decreasing trends of suspended particulate matter and PM  $_{2.5}$  concentrations in Tokyo, 1990–2010. Journal of the Air & Waste Management Association 63, 737–748. <https://doi.org/10.1080/10962247.2013.782372>
- [19] Henze, D.K., Hakami, A., Seinfeld, J.H., 2007. Development of the adjoint of GEOS-Chem. Atmospheric Chemistry and Physics 7, 2413–2433. [https://doi.org/10.5194/acp-7-2413-](https://doi.org/10.5194/acp-7-2413-2007) [2007](https://doi.org/10.5194/acp-7-2413-2007)
- [20] Hoesly, R.M., Smith, S.J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J.J., Vu, L., Andres, R.J., Bolt, R.M., Bond, T.C., Dawidowski, L., Kholod, N., Kurokawa, J., Li, M., Liu, L., Lu, Z., Moura, M.C.P., O'Rourke, P.R., Zhang, Q., 2018. Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS). Geoscientific Model Development 11, 369–408.<https://doi.org/10.5194/gmd-11-369-2018>
- [21] Hu, J., Huang, L., Chen, M., Liao, H., Zhang, H., Wang, S., Zhang, Q., Ying, Q., 2017. Premature Mortality Attributable to Particulate Matter in China: Source Contributions and Responses to Reductions. Environmental Science & Technology 51, 9950–9959. <https://doi.org/10.1021/acs.est.7b03193>
- [22] Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J.J.P., Klimont, Z., Frost, G., Darras, S., Koffi, B., Li, M., 2015. HTAP\_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution. Atmospheric Chemistry and Physics 15, 11411–11432. <https://doi.org/10.5194/acp-15-11411-2015>
- [23] Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., Schöpp, W., 2017. Global anthropogenic emissions of particulate matter including black carbon. Atmospheric Chemistry and Physics 17, 8681–8723. <https://doi.org/10.5194/acp-17-8681-2017>
- [24] Krewski, D., Jerrett, M., Burnett, R.T., Ma, R., Hughes, E., Shi, Y., Turner, M.C., Pope III, C.A., Thurston, G., Calle, E.E., others, 2009. Extended follow-up and spatial analysis of the American Cancer Society study linking particulate air pollution and mortality. Health Effects Institute Boston, MA. URL: [https://www.healtheffects.org/publication/extended](https://www.healtheffects.org/publication/extended-follow-and-spatial-analysis-american-cancer-society-study-linking-particulate)[follow-and-spatial-analysis-american-cancer-society-study-linking-particulate](https://www.healtheffects.org/publication/extended-follow-and-spatial-analysis-american-cancer-society-study-linking-particulate)
- [25] Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., Akimoto, H., 2013. Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in Asia (REAS) version 2. Atmospheric Chemistry and Physics 13, 11019–11058. <https://doi.org/10.5194/acp-13-11019-2013>
- [26] Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D., Pozzer, A., 2015. The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature 525, 367– 371.<https://doi.org/10.1038/nature15371>
- [27] Lewis, C.A., 1997. Fuel and Energy Production Factors, MEET Project (Methodologies for Estimating Air Pollutant Emissions from Transport) funded by the European Commission under the Transport RTD program of the 4<sup>th</sup> framework. European Commission. URL: [http://citeseerx.ist.psu.edu/viewdoc/download?doi=10.1.1.507.7246&rep=rep1&type=](http://citeseerx.ist.psu.edu/viewdoc/download?doi=10.1.1.507.7246&rep=rep1&type=pdf) [pdf](http://citeseerx.ist.psu.edu/viewdoc/download?doi=10.1.1.507.7246&rep=rep1&type=pdf)
- [28] Li, M., Zhang, Q., Kurokawa, J., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D.G., Carmichael, G.R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., Zheng, B., 2017. MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP. Atmospheric Chemistry and Physics 17, 935–963.<https://doi.org/10.5194/acp-17-935-2017>
- [29] Li, M., Zhang, Q., Streets, D.G., He, K.B., Cheng, Y.F., Emmons, L.K., Huo, H., Kang, S.C., Lu, Z., Shao, M., Su, H., Yu, X., Zhang, Y., 2014. Mapping Asian anthropogenic emissions of non-methane volatile organic compounds to multiple chemical mechanisms. Atmospheric Chemistry and Physics 14, 5617–5638. [https://doi.org/10.5194/acp-14-](https://doi.org/10.5194/acp-14-5617-2014) [5617-2014](https://doi.org/10.5194/acp-14-5617-2014)
- [30] Liu, F., Beirle, S., Zhang, Q., van der A, R.J., Zheng, B., Tong, D., He, K., 2017. NO<sub>x</sub> emission trends over Chinese cities estimated from OMI observations during 2005 to 2015. Atmospheric Chemistry and Physics 17, 9261–9275. [https://doi.org/10.5194/acp-](https://doi.org/10.5194/acp-17-9261-2017)[17-9261-2017](https://doi.org/10.5194/acp-17-9261-2017)
- [31] Liu, F., Zhang, Q., Tong, D., Zheng, B., Li, M., Huo, H., He, K.B., 2015. High-resolution inventory of technologies, activities, and emissions of coal-fired power plants in China from 1990 to 2010. Atmospheric Chemistry and Physics 15, 13299–13317. <https://doi.org/10.5194/acp-15-13299-2015>
- [32] Liu, F., Zhang, Q., van der A, R.J., Zheng, B., Tong, D., Yan, L., Zheng, Y., He, K., 2016. Recent reduction in  $NO<sub>x</sub>$  emissions over China: synthesis of satellite observations and emission inventories. Environmental Research Letters 11, 114002. <https://doi.org/10.1088/1748-9326/11/11/114002>
- [33] Lozano, R. et al., 2012. Global and regional mortality from 235 causes of death for 20 age groups in 1990 and 2010: a systematic analysis for the Global Burden of Disease Study 2010. The Lancet 380, 2095–2128. [https://doi.org/10.1016/S0140-](https://doi.org/10.1016/S0140-6736(12)61728-0) [6736\(12\)61728-0](https://doi.org/10.1016/S0140-6736(12)61728-0)
- [34] Nayak, C.K., Jadhav, R., 2012. Govt cuts subsidy on most fertilizers for 2012/13. Reuters. URL: [https://in.reuters.com/article/india-fertiliser-subsidy/govt-cuts-subsidy-on-most](https://in.reuters.com/article/india-fertiliser-subsidy/govt-cuts-subsidy-on-most-fertilizers-for-2012-13-idINDEE8200AD20120301)[fertilizers-for-2012-13-idINDEE8200AD20120301](https://in.reuters.com/article/india-fertiliser-subsidy/govt-cuts-subsidy-on-most-fertilizers-for-2012-13-idINDEE8200AD20120301)
- [35] National Bureau of Statistics of China, 2017. China Statistical Yearbook 2017. National Bureau of Statistics of China. URL:<http://www.stats.gov.cn/tjsj/ndsj/2017/indexeh.htm>
- [36] NASA Earth Observations, 2018. Aerosol Optical Thickness (1 Month). National Aeronautics and Space Administration. URL: https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MODAL2 M\_AER\_OD&year=2010
- [37] ORNL, 2010. LandScan 2010 High-Resolution Global Population Data Set. Oak Ridge National Laboratory. URL:<https://landscan.ornl.gov/>
- [38] ORNL, 2016. LandScan 2015 High-Resolution Global Population Data Set. Oak Ridge National Laboratory. URL:<https://landscan.ornl.gov/>
- [39] Randerson, J.T., van der Werf, G.R., Giglio, L., Collatz, G.J., Kasibhatla, P.S., 2017. Global Fire Emissions Database, Version 4.1 (GFEDv4). <https://doi.org/10.3334/ORNLDAAC/1293>
- [40] Silva, R.A., West, J.J., Zhang, Y., Anenberg, S.C., Lamarque, J.-F., Shindell, D.T., Collins, W.J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L.W., Nagashima, T., Naik, V., Rumbold, S., Skeie, R., Sudo, K., Takemura, T., Bergmann, D., Cameron-Smith, P., Cionni, I., Doherty, R.M., Eyring, V., Josse, B., MacKenzie, I.A., Plummer, D., Righi, M., Stevenson, D.S., Strode, S., Szopa, S., Zeng, G., 2013. Global premature mortality due to anthropogenic outdoor air pollution and the contribution of past climate change. Environmental Research Letters 8, 034005. [https://doi.org/10.1088/1748-](https://doi.org/10.1088/1748-9326/8/3/034005) [9326/8/3/034005](https://doi.org/10.1088/1748-9326/8/3/034005)
- [41] Soares, C.G., Santos, T.A., 2015. Maritime technology and engineering: proceedings of MARTECH 2014, 2nd International Conference on Maritime Technology and Engineering, Lisbon, Portugal, 15-17 October 2014.
- [42] Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S.D., Fu, Q., He, D., Klimont, Z., Nelson, S.M., Tsai, N.Y., Wang, M.Q., Woo, J.-H., Yarber, K.F., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000: AEROSOL EMISSION INVENTORY. Journal of Geophysical Research: Atmospheres 108. <https://doi.org/10.1029/2002JD003093>
- [43] Tokyo Metropolitan Government, 2018. Air Quality Control: PM<sub>2.5</sub>. Tokyo Metropolitan Government Bureau of Environment. [http://www.kankyo.metro.tokyo.jp/en/automobile/pm25\\_monitoring.html](http://www.kankyo.metro.tokyo.jp/en/automobile/pm25_monitoring.html)
- [44] Tong, D., Zhang, Q., Davis, S.J., Liu, F., Zheng, B., Geng, G., Xue, T., Li, M., Hong, C., Lu, Z., Streets, D.G., Guan, D., He, K., 2018. Targeted emission reductions from global superpolluting power plant units. Nature Sustainability 1, 59–68. <https://doi.org/10.1038/s41893-017-0003-y>
- [45] UNPD, 2014. World Urbanization Prospects: 2014 Revision. United Nations Population Division. URL:<https://data.worldbank.org/indicator/SP.URB.TOTL.IN.ZS>
- [46] UNSD, 2018. Guidelines for the 2016 United Nations Statistics Division Annual Questionnaire on Energy Statistics. United Nations Statistics Division. URL: <https://unstats.un.org/unsd/energy/Energy-Questionnaire-Guidelines.pdf>
- [47] U.S. EPA, 1995. AP 42, Fifth Edition Compilation of Air Pollutant Emissions Factors, Volume 1: Stationary Point and Area Sources. United States Environmental Protection Agency. URL: [https://www.epa.gov/air-emissions-factors-and-quantification/ap-42](https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emissions-factors) [compilation-air-emissions-factors](https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emissions-factors)
- [48] U.S. EPA, 2015. 2011 National Emissions Inventory, Technical Support Document. United States Environmental Protection Agency. URL: [https://www.epa.gov/air-emissions](https://www.epa.gov/air-emissions-inventories/2011-national-emissions-inventory-nei-technical-support-document)[inventories/2011-national-emissions-inventory-nei-technical-support-document](https://www.epa.gov/air-emissions-inventories/2011-national-emissions-inventory-nei-technical-support-document)
- [49] U.S. EPA, 2016. 2014 National Emissions Inventory, Technical Support Document. United States Environmental Protection Agency. URL: [https://www.epa.gov/air-emissions](https://www.epa.gov/air-emissions-inventories/2014-national-emissions-inventory-nei-technical-support-document-tsd)[inventories/2014-national-emissions-inventory-nei-technical-support-document-tsd](https://www.epa.gov/air-emissions-inventories/2014-national-emissions-inventory-nei-technical-support-document-tsd)
- [50] USGS, 2018. USGS National Minerals Information Center: Commodity Statistics and Information. United States Geological Survey. URL: <https://minerals.usgs.gov/minerals/pubs/commodity/>
- [51] Wang, H. et al., 2016. Global, regional, and national life expectancy, all-cause mortality, and cause-specific mortality for 249 causes of death, 1980–2015: a systematic analysis for the Global Burden of Disease Study 2015. The Lancet 388, 1459–1544. [https://doi.org/10.1016/S0140-6736\(16\)31012-1](https://doi.org/10.1016/S0140-6736(16)31012-1)
- [52] Winnes, H., Fridell, E., 2009. Particle Emissions from Ships: Dependence on Fuel Type. Journal of the Air & Waste Management Association 59, 1391–1398. <https://doi.org/10.3155/1047-3289.59.12.1391>
- [53] Xu, Y., Hu, J., Ying, Q., Hao, H., Wang, D., Zhang, H., 2017. Current and future emissions of primary pollutants from coal-fired power plants in Shaanxi, China. Science of the Total Environment 595, 505–514.<https://doi.org/10.1016/j.scitotenv.2017.03.267>
- [54] Zender, C.S., 2003. Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology. Journal of Geophysical Research 108. <https://doi.org/10.1029/2002JD002775>
- [55] Zhang, Q., Streets, D.G., Carmichael, G.R., He, K.B., Huo, H., Kannari, A., Klimont, Z., Park, I.S., Reddy, S., Fu, J.S., Chen, D., Duan, L., Lei, Y., Wang, L.T., Yao, Z.L., 2009. Asian emissions in 2006 for the NASA INTEX-B mission. Atmospheric Chemistry and Physics 9, 5131–5153.<https://doi.org/10.5194/acp-9-5131-2009>
- [56] Zhao, Y., Wang, S., Nielsen, C.P., Li, X., Hao, J., 2010. Establishment of a database of emission factors for atmospheric pollutants from Chinese coal-fired power plants.

Atmospheric Environment 44, 1515–1523. <https://doi.org/10.1016/j.atmosenv.2010.01.017>

- [57] Zheng, B., Huo, H., Zhang, Q., Yao, Z.L., Wang, X.T., Yang, X.F., Liu, H., He, K.B., 2014. High-resolution mapping of vehicle emissions in China in 2008. Atmospheric Chemistry and Physics 14, 9787–9805.<https://doi.org/10.5194/acp-14-9787-2014>
- [58] Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., Zhang, Q., 2018. Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. Atmospheric Chemistry and Physics Discussions 1–27.<https://doi.org/10.5194/acp-2018-374>