Where Does Ambient $PM_{2.5}$ Come from in China: A Comparison and Synthesis of Recent Estimates

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Abstract

Exposure to ambient $PM_{2.5}$ in China is reported as the 3rd largest contributor to excess deaths in China (approximately 1.1 million annually). The breakdown of pollutant sources to $PM_{2.5}$ exposure would help policymakers prioritize pollution control strategies, but recent studies on this topic (*GBD MAPS Working Group, 2016; Hu et al., 2017a; Karagulian et al., 2017; Lelieveld et al., 2015; Silva et al., 2016*) greatly diverge. We systematically inter-compare meteorological data, model resolution, aerosol chemistry and emission inventories that applied in these studies to better understand the causes of the divergent conclusions. We find that the divergent conclusions can be explained well by the definition of sector categories and statistical characteristics of these inventories. We synthesize the results from studies using similar sector categories and based on more recent years, and conclude that industrial sector (including energy manufacturing) dominates the sector contribution (32%, 28-37%) to population-weighted $PM_{2.5}$ in China, and needs immediate action.

1 Introduction

The Chinese economy has been undergoing rapid changes since the economic reforms began in 1978, with the gross domestic product (GDP) in 2017 about 60 times of that in 1978 (*National Bureau of Statistics of China，2018*). Such a dramatic economic development has stimulated an increase in energy consumption and has led to hazardous air pollution. In recent decades, frequent haze episodes in China with extremely high concentrations of $PM_{2.5}$ were prominently featured in global news and media channels, and received worldwide attention. $PM_{2.5}$ pollution has been linked to visibility reduction (*Gao et al., 2016*), climate change (*Menon et al., 2002*), ecological degradation (*Unger et al., 2017*), and most importantly, human health risks (*West et al., 2016*). $PM_{2.5}$ particles are small enough to enter deep into the lungs to cause cardiovascular diseases, as well as chronic pulmonary disease, lung cancer, and childhood pneumonia (*Pope III and Dockery, 2006*). Exposure to ambient $PM_{2.5}$ in China was estimated to cause approximately 1.1 million deaths annually, making it the 3rd largest contributor to deaths in China (*Cohen et al., 2017*).

The public discontent over $PM_{2.5}$ pollution in China has prompted the government to take

action against it since 2013 (*Zheng et al, 2018*). However, limited and uncertain understanding of source contribution to $PM_{2.5}$ confuses policymakers and hinders the effectiveness of control strategies. Ambient $PM_{2.5}$ comes from various sources, including power generation, industry, residential, transportation, biomass burning, etc., through direct emission and/or secondary aerosol formation (*Liu et al, 2016a*). Understanding the relative importance of these emission sectors to $PM_{2.5}$ exposure can help the government prioritize pollution control policies in China.

With the advances in $PM_{2.5}$ measurements, modeling, and statistical methods to merge different $PM_{2.5}$ datasets (*Gao et al., 2017; Liang et al., 2017*), the uncertainties in estimating $PM_{2.5}$ exposure have been reduced, and estimates of mortality attributable to $PM_{2.5}$ in China from recent studies are generally consistent (*Archer-Nicholls et al., 2016; Lelieveld et al., 2015; Liu et al., 2016b*). Nevertheless, results from limited studies on the breakdown of pollutant sources to $PM_{2.5}$ exposure in China differ greatly.

Until now, there have been five published studies (*GBD MAPS Working Group, 2016; Hu et al., 2017a; Karagulian et al., 2017; Lelieveld et al., 2015; Silva et al., 2016*) comprehensively estimating the contributions of major source sectors to population-weighted $PM_{2.5}$ globally and for China. However, rather wide ranges are seen in their reported contributions. It is challenging to interpret these results as different chemical transport models, emission inventories, base year, and other assumptions were used. In this study, we systematically compare the methodology applied in these studies, discuss the causes of the differences in conclusions, and reconcile the results to provide better estimates as references for policymakers.

2 Inter-comparison of the Methodology and Sector Contributions

The above mentioned five modeling studies (referred as studies A-E in Table 1; *GBD MAPS Working Group, 2016; Hu et al., 2017a; Karagulian et al., 2017; Lelieveld et al., 2015; Silva et al., 2016*) use global or regional air quality models, along with emission inventories with source-sector information to apportion $PM_{2.5}$ exposure to source sectors. Unlike the common conclusion among a recent review of several Indian studies (*Chowdhury et al., 2018; Conibear*

et al., 2018) that residential sector is the dominant source, large disagreements of the relative importance of emission sectors are found for China. Studies A and B reveal that residential sector dominates the contribution to population-weighted $PM_{2.5}$ in China, while studies C-E conclude that industrial sector plays an even more important role (Table 1). Such large disagreements in conclusions impede effective policymaking with regard to: (1) which source sector should be prioritized for control; (2) by what extent the control strategies can reduce ambient $PM_{2.5}$ exposure. To better interpret the above highly heterogeneous results, it is crucial to understand why they are different. In this section, we scrutinize the methodology adopted by each study and examine how it is associated with the results. Table 2 summarizes the characteristics of modeling frameworks applied in studies A-E.

2.1 Meteorological data

Meteorological processes influence $PM_{2.5}$ concentrations strongly through advection, diffusion, vertical transport, gas and aqueous-phase chemistry, deposition, etc. Meteorological fields in studies A and C are either driven or adopted nudging correction by ECMWF (European Centre for Medium-Range Weather Forecasts, https://www.ecmwf.int/) analyses, while both studies B and D take meteorological fields from the NASA GEOS (Goddard Earth Observing System) analyses. These global meteorological analyses datasets embody the information from meteorological observations, so they are likely to accurately represent large scale and synoptic scale meteorology. However, assumptions in the large scale meteorological models might lead to information incompleteness when applied to regional or local studies, such as sub-grid scale cloud dynamics, complex topographical features, and boundary layer processes. Study E uses a regional scale weather model WRF (Weather Research and Forecasting) with 36km×36km grid size to simulate meteorology fields. The settings are expected to be applicable to the China situations, and the capabilities of this model in simulating meteorological features in China have been shown in a variety of peer-reviewed publications (*Gao et al., 2016; Hu et al., 2016*).

2.2 Model resolution, Aerosol chemistry and Model Evaluation

Global models used in studies A and C apply grid sizes larger than 1 degree, while other studies (B, D and E) adopt grid size of ~50km or less (Table 2). Previous studies have appreciated the

importance of air quality model resolution on uncertainty associated with health impacts (*Kushta et al., 2018; Li et al., 2016; Punger and West, 2013;*). Both studies *Li et al. (2016)* and *Punger and West (2013)* found that coarse grid resolutions tend to estimate biased low for $PM_{2.5}$ exposure (<20% lower for resolutions <100km). However, *Kushta et al. (2018)* concluded that model resolution is a relatively small factor in the uncertainty (estimates using 20km resolution are 2.4% higher than results from 100km resolution).

Reactions parameterized in chemical transport models have direct influences on $PM_{2.5}$ simulations (*Gao et al., 2018*). The EMAC model in study A considers all important aerosol components in the atmosphere, and the secondary organic aerosol (SOA) is treated according to *Tsimpidi et al. (2014)*, linked to the particle microphysics module of *Pringle et al. (2010*). The thermodynamic equilibrium model ISORROPIA-II was incorporated into EMAC to calculate gas/aerosol partitioning (*Fountoukis and Nenes, 2007*). The MOZART-4 model in study B uses simple approximations to determine the amounts of ammonium nitrate, and SOA formation are formed from the oxidation of monoterpenes and toluene (*Emmons et al., 2010*). The treatments of aerosols in the TM5-FASST model is comparable to EMAC (*Van Dingenen et al., 2015, 2018*). The GEOS-Chem model (study D) includes SOA from more pathways than other global models, such as the reaction of isoprene with reactive nitrogen (*Pye et al., 2010*). CMAQ (study E) considers also sophisticated parameterizations of SOA, including oxidation of both anthropogenic and biogenic sources (*Hu et al., 2017b*).

Studies A-C are global model studies with no validation of $PM_{2.5}$ concentrations for China. Studies D and E compare model $PM_{2.5}$ against nationwide settled observations, and reveal that annual mean $PM_{2.5}$ concentrations are generally underestimated by both models, which might lead to the underappreciation of mortality attributable to $PM_{2.5}$.

2.3 Emission inventories

Estimating emissions are challenging, particularly in developing counties with less data constraints. The existing errors in emission estimates, along with discrepancies between inventories compiled at different institutions are expected to lead to notable dissimilarities in simulated $PM_{2.5}$ concentrations, estimated $PM_{2.5}$ exposure and the relative importance of individual source sector.

EDGAR-HTAP (Emissions Database for Global Atmospheric Research-Hemispheric Transport of Air Pollution, *Janssens-Maenhout et al., 2012*), RCP (Representative Concentration Pathways) 8.5 (*Lamarque et al., 2010*), HTAP V2 (*Janssens-Maenhout et al., 2015*), School of Environment of Tsinghua University (SOE) (*Ma et al., 2017*; *Wang et al., 2011*), and the Multi-resolution Emission Inventory for China (MEIC, *Li et al., 2017*) inventories were used by studies A-E, respectively. These inventories consider anthropogenic emissions of primary aerosols and aerosol precursors (SO_2, NO_x, NMVOCs, NH_3, BC, OC, $PM_{2.5}$, and PM_{10}). Table 3 summarizes the annual emissions of anthropogenic species and sectoral contributions in China from these inventories.

The amount of SO_2 estimated in these inventories are generally consistent except that the SOE inventory (study D) are lower than others. This is the SOE inventory was developed for year 2013, while other inventories are based on years earlier than that. Strict SO_2 control measures implanted in China might have led to this gap. According to the statistics in *Zheng et al. (2018)*, SO_2 emissions in China declined by 9% from 2010 to 2013. Energy/power and industrial sectors are the largest contributors to SO_2 emissions. However, the EDGAR-HTAP and RCP 8.5 inventories (studies A and B) exhibit larger contribution from energy/power sector and less contribution from industrial sector, compared to other inventories. This is related to the sector aggregation during the developments of these inventories. In the EDGAR-HTAP and RCP 8.5 inventories (studies A and B), manufacture of solid fuels and other energy industries are included in energy sector instead of industrial sector, leading to higher contribution of energy sector and lower contribution of industrial sector. Although in the HTAP V2 inventory, the terminology energy is used, only power generation is included (*Janssens-Maenhout et al., 2015*).

The total amount of NO_x emissions in the EDGAR-HTAP and RCP 8.5 inventories (studies A and B) are much lower than other estimates. The statistics of the EDGAR-HTAP and RCP 8.5 inventories (studies A and B) shown in Table 3 are based on year 2005, and there were dramatic enhancements in NO_x emissions, particularly from transportation sector, during 2004-2013 (*Zhang et al., 2007*; *Zheng et al., 2018*). The amount of NH_3 emissions from the EDGAR-HTAP inventory (study A) is notably higher than other estimates (32% higher than the mean of NH_3 emissions used in studies B, C and D). Such high estimates of NH_3 emissions in study

A might be one of the reasons for higher attributed contribution from agricultural sector to mortality (Table 1).

The estimates of total emissions of BC, OC, $PM_{2.5}$, and PM_{10} from these inventories are generally consistent, although the sectoral contributions greatly differ. Based on the statistics of the inventories used in studies C-E, industrial sector is the largest contributor to primary $PM_{2.5}$ emissions, while the inventory used in A (EDGAR-HTAP) shows higher contribution from residential sector. The RCP 8.5 inventory (study B) does not consider emissions of primary $PM_{2.5}$ except BC and OC (road dust, etc.), which could be the reason for the conclusion drew from studies A and B that residential sector is the dominant sector instead of industrial sector in China.

The estimated contribution of transportation sector is generally below 10% except in study D. Previous studies show also that reduction in transportation sector exhibits less importance than power and industry, and residential sectors in terms of contribution to population-weighted $PM_{2.5}$ in East Asia (*Liang et al., 2018*), and the share of transportation sector in East Asia population-weighted $PM_{2.5}$ is only about 5% (*Chambliss et al., 2014*). We compared the transportation emissions in the SOE and MEIC inventories (studies D and E), and found that transportation generated SO_2 emissions in the SOE inventory is four times of that in MEIC (Table 3). Such high SO_2 emissions can lead to higher estimated contribution of transportation sector in study D.

The estimated contribution from residential sector range from 19% to 32%, with relatively higher estimates from studies A and B. We compared estimates of residential contribution from A-E against other studies focusing on residential sector: *Chaff et al. (2016)* (10%) and *Archer-Nicholls et al. (2016)* (37%). *Chaff et al. (2016)* only considers household cooking stoves and open-pit cooking emissions, while the emissions used in *Archer-Nicholls et al. (2016)* are inclusive of commercial emissions, as well as household emissions for cooking, heating and lighting. The use of the GBD-related databases (http://www.healthdata.org/gbd) for determining emissions from household solid fuel use underestimates the total emissions from households, since it does not include non-cooking uses, particularly in the winter and in the north.

3.3 Health Calculation

The contributions from different sectors in Table 1 to $PM_{2.5}$ exposure are presented in different ways. Studies B, C and D calculate source contributions to population-weighted $PM_{2.5}$ concentrations, which were conducted by zeroing out each sector in the simulations, and estimated using the following two equation:

$$PM_{2.5,s} = PM_{BASE} - PM_{zero-s} \quad (1)$$

$$PWPM_{2.5,s} = \frac{\sum_j PM_{2.5,s} \times N_j}{\sum_j N_j} \quad (2)$$

where PM_{BASE} and PM_{zero-s} denote annual mean surface $PM_{2.5}$ concentrations from the BASE simulation (with all emissions sectors) and zero-s (zeroing out the emissions of sector s) cases. N_j represents the population in gridcell j. $PM_{2.5,s}$ is taken as the sector s induced $PM_{2.5}$ concentrations, and $PWPM_{2.5,s}$ stands for the population-weighted $PM_{2.5}$ concentrations. $PM_{2.5,s}$ in equation (2) can be replaced by PM_{BASE} to calculate total population-weighted $PM_{2.5}$ ($PWPM_{2.5}$), and the ratio of $PWPM_{2.5,s}$ to $PWPM_{2.5}$ serves as the factional contribution of sector s to $PM_{2.5}$ exposure.

Nevertheless, a special approach was adopted by *Lelieveld et al. (2015)*, in which the contribution of a source class was computed using:

$$F_s = \frac{M(PM_{BASE}) - M(PM_{zero-s})}{M(PM_{BASE})} \quad (3)$$

where M stands for excess mortality.

These two approaches differ in the question they seek to answer. The approach of *Lelieveld et al. (2015)* is guided by the tradition of forward-looking 'consequential' analysis, which seeks to determine how large of a reduction in mortality would be expected if emissions from a single source class were eliminated. In contrast, the approach taken by studies B-D follows the backward-looking 'attributional' analysis that attempts to determine what fraction of total $PM_{2.5}$ related mortality is caused by emissions of a single source. When the exposure-response function is linear, answers from these two approaches would not conflict. But given the strong nonlinearities of the IER concentration-response function, remarkable dissimilarities are found in Table 1. In view of current high levels of $PM_{2.5}$ concentrations in China, the forward-looking 'consequential' approach adopted by *Lelieveld et al. (2015)* is likely to show substantially smaller estimates of source class impacts than those from the attributional approach.

Given the non-linearity of aerosol chemistry, the sector elimination method adopted by studies A-D can cause non-conservation of sector contributions, although it does not affect the interpretation of relative importance. For example, the sum of sector contributions considered in studies B and C are less than 100%. Study E use a source-oriented version of chemical transport model, which can overcome the non-conservation limitation. The calculation of source contribution to mortality is same as studies B-D also follows the backward-looking 'attributional analysis' approach.

3.4 Synthesis of Recent Estimates

According to the above discussion on the influences of meteorological data, model resolution, aerosol chemistry, and emission inventory, large difference in the used inventories is likely to be the major reason for the discrepancies in the conclusions from the discussed five studies. Given the definition of emission categories in studies A and B are different from those in studies C-E, and the based year for studies C-E are more recent, we synthesize the results from studies C-E here. The mean contribution of each sector is shown in Figure 1. Industrial sector dominates the sector contribution (32%, 28-37%) to population-weighted $PM_{2.5}$ in China, followed by residential sector (23%, 19-27%). Agricultural (12%), power generation (12%, 10-16%), and transportation (9%, 6-15%) play relatively less important roles.

3 Discussion and Conclusion

To date, there have been five global or national studies comprehensively evaluating the contribution of source sectors to $PM_{2.5}$ exposure in China. However, the conclusions greatly differ, which confuses policymakers and hinders the effectiveness of control strategies. Here, we inter-compare meteorological data, model resolution, aerosol chemistry and emission inventories that applied in these studies to better understand the causes of the divergent conclusions.

Most studies used meteorological data from reanalysis datasets with observational constraints that can accurately represent large scale and synoptic scale meteorology. Previous sensitivity studies (*Kushta et al., 2018*) point out that model resolution is a relatively minor factor in the uncertainty of $PM_{2.5}$ exposure estimation. In general, national scale studies (D and E) are

expected to provide better estimates with optimized model configurations (model resolution, meteorological data, emission inventories, etc.) for China, and more thorough evaluation of simulation results considering the availability of surface $PM_{2.5}$ measurements. Global models have the strengths of simulating transboundary phenomena, although the contribution of transboundary pollution imports into China is expected to be of minor significance (*Dong et al., 2018*) given the short lifetime of aerosols and the large emission strengths of local sources. The treatments of aerosol chemistry in these studies are generally similar, except that national studies include more detailed treatments of SOA.

Through detailed comparisons of the emission inventories (HTAP-EDGAR, RCP 8.5, HTAP V2, SOE, and MEIC) used in this studies, we find that the divergent conclusions can be explained well by the definition of sector categories and statistical characteristics of these inventories. In the EDGAR-HTAP and RCP 8.5 inventories, manufacture of solid fuels and other energy industries are included in energy sector instead of industrial sector, which are considered in industrial sector in other inventories. Total NH_3 emissions in EDGAR-HTAP are nearly 1.3 times that of the SOE inventory, which could be one of the reasons for a notably high contribution by the agricultural sector in China inferred by *Lelieveld et al. (2015)*. Given the differences in definitions of sectors and relatively old based year in studies A and B, we synthesize the results from studies C-E. Industrial sector (including energy manufacturing) dominates the sector contribution (32%, 28-37%) to population-weighted $PM_{2.5}$ in China, and needs immediate action.

These results represent the conditions during 2010-2013, and conclusion might change as these inventories become out of date. Since 2013, the Chinese government implemented stricter control policies, including improving fuel quality, retiring old vehicles, eliminating small coal-fired boilers, phasing out small and polluted factories, etc. (*Zheng et al., 2018*). Due to China's Clean Air Action, anthropogenic emissions were estimated to decrease by 59% for SO_2, 21% for NO_x, 28% for BC, 32% for OC, 33% for $PM_{2.5}$ and 11% for NMVOCs during 2013-2017 period (*Zheng et al., 2018*). Future concentrated control measures need reassessments based on the most recent emission conditions.

Given certain discrepancies found in this study come from the definition of emission categories, future developments of inventories should give a clear breakdown of the defined emission

sectors, with some attempt to coordinate it internationally. Besides, most studies discussed here do not use local data in China to validate model results, partially due to the inaccessibility to measurement data sets. Establishing publicly available measurement datasets would facilitate the understanding of sources and uncertainties in China. The results in most studies are presented at country scale and are based on annul mean, which may have limited policy implications, since controlling policies are usually at local and provincial level. Providing more detailed information at higher spatial and temporal scales in the future would be helpful.

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Table 1 Contribution of source sectors to population-weighted $PM_{2.5}$ for studies A-E (%)

Studies	A *Lelieveld et al. 2015*	B *Silva et al., 2016*	C *Karagulian et al., 2017*	D *GBD MAPS Working Group, 2016*	E *Hu et al., 2017a*
Power/Energy	17	17	16	10	10
Residential	28	32	27	19	23
Industrial	9	26	37	28	31
Transportation	3	8	6	15	6
Agricultural	27	n/a	12	n/a	12
Open burning	1	n/a	3	7	4
Natural	15	9	n/a	n/a	n/a

Table 2 Characteristics of the adopted modeling frameworks

	A	B	C	D	E
Year	2010	2005	2010	2013	2013
Model	EMAC	MOZART-4	TM5-FASST	GEOS-Chem	WRF-CMAQ
Resolution	1.1°× 1.1°	0.67°× 0.5°	1°× 1° (1°×1° over most of the source regions, and downgraded to 3°×2° and 6°×4° away from the continents)	0.67°× 0.5°	36km× 36km
Aerosols	SO_4^{2-}, NO_3^-, NH_4^+, BC, POM, dust, and sea salt; 15% of natural terpene emissions form SOA (*Pringle et al., 2010*)	SO_4^{2-}, BC, POM, NH_4NO_3 (simple approximations) dust, and sea salt; SOA: oxidation of monoterpenes and toluene (*Emmons et al., 2010*)	SO_4^{2-}, NO_3^-, NH_4^+, BC, POM, dust, sea salt and other primary $PM_{2.5}$; 15% of natural terpene emissions form SOA (*Van Dingenen et al., 2015, 2018*)	SO_4^{2-}, NO_3^-, NH_4^+, BC, POM, dust, and sea salt; SOA: oxidation of monoterpenes, toluene, isoprene and aromatics (*Heald et al., 2014*)	SO_4^{2-}, NO_3^-, NH_4^+, BC, POM, dust, soil, and sea salt; SOA: oxidation of several VOCs (anthropogenic + biogenic) and two-product approach (*Hu et al., 2017b*)
Inventory	EDGAR-HTAP	RCP 8.5 global emissions for 2005	HTAP V2	SOE inventory for China	MEIC 2012 for China

Table 3 Anthropogenic emissions and sectoral contributions (Tg/yr)

	Studies	SO_2	NO_x	NMVOCs	NH_3	BC	OC	$PM_{2.5}$	PM_{10}
Total	A	31.5	16.9	23.8	12.5	1.4	2.8	12.7	17.9
	B	28.4	18	27.0	7.0	1.6	3.6	NA	NA
	C	28.6	29	23.6	9.1	1.8	3.4	12.2	16.6
	D	23.2	25.6	23.4	9.6	2.0	3.4	12.2	16.5
	E	28.5	29.2	28.1	10.7	1.8	3.2	11.9	16.5
ENE	A	27.7	12.2	0.9	0.0	0.3	0.3	5.2	8.5
	B	18.5	6.5	1.0	0.0	0.0	0.1	NA	NA
	C	8.1	9.4	0.3	0.0	0.0	0.0	0.9	1.4
	D	6.3	6.5	0.0	0.0	0.0	0.0	0.6	1.0
	E	6.9	9.1	0.1	0.0	0.0	0.0	0.9	1.3
RES	A	2.2	1.6	11.3	0.0	0.7	1.9	4.7	5.1
	B	3	1.8	13.8	0.1	0.6	2.1	NA	NA
	C	3.5	1.2	6.3	0.2	0.9	2.8	4.7	5.2
	D	3	1.1	4.3	0.9	0.9	1.9	3.9	4.3
	E	3.7	1.1	5.0	0.4	0.9	2.5	4.4	4.9
TRA	A	0.3	3	2.4	0.0	0.1	0.2	0.3	0.3
	B	0.7	2.5	4.2	0.0	0.1	0.1	NA	NA
	C	0.2	7.2	2.5	0.1	0.3	0.1	0.5	0.5
	D	0.9	7.2	2.9	0.0	0.2	0.1	0.6	1.0
	E	0.3	8.5	5.6	0.0	0.3	0.1	0.5	0.5
IND	A	1	11.7	6.6	0.0	0.0	0.1	1.1	1.6
	B	6.1	6.5	2.2	0.0	0.8	1.2	NA	NA
	C	16.8	11.2	14.5	0.2	0.6	0.5	6.1	9.5
	D	12.9	10.3	6.4	0.2	0.8	0.6	5.9	9.1
	E	17.6	10.5	8.9	0.3	0.6	0.6	6.1	9.7
AGR	A				11.5				
	B				6.8				
	C				8.7				
	D				8.5				
	E				9.9				

ENE: energy sector in A and B (including fossil fuel manufacturing industries), power generation in C, D and E; RES: residential sector; TRA: transportation sector; IND: industrial sector; AGR: agricultural sector (A includes waste burning)

Statistics shown in the table are based on year 2005 (A), 2005 (B), 2010 (C), 2013 (D) and 2012 (E).

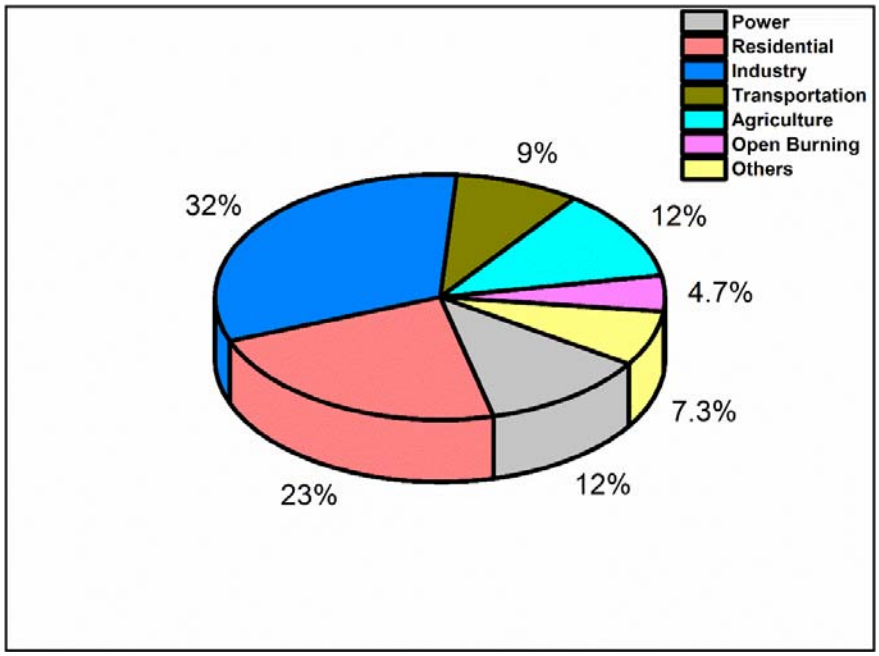

Figure 1 Contribution of emission sectors to population-weighted $PM_{2.5}$ in China