

Original Research

# Atmospheric Emissions of As, Sb, and Se from Coal Combustion in Shandong Province, 2005-2014

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## Abstract

The emissions of hazardous trace elements have gained considerable attention because of their negative impacts on local air quality, regional environmental health, and ecological risks. Shandong Province has been considered to be the top provincial emitter of Sb, As, and Se in China owing to rapid economic development and its energy consumption structure (mainly coal). In this study we investigate the atmospheric emissions of Sb, As, and Se from coal combustion in Shandong from 2005 to 2014, and we analyze a scenario for future emissions from coal-fired power plants. The inventory is based on the following parameters: coal consumption, economic sectors, boiler types, and air pollution control technologies. Results indicate that the calculated provincial total emissions of Sb, As, and Se from coal combustion in 2005 were estimated at 40.26, 246.5, and 255.9 t, respectively, and increased to 51.36, 311.9, and 313.9 t by 2014 with annual growth rates of 2.75%, 2.65%, and 2.27%. Industrial use was the largest single sector, accounting for nearly 83.2%, 82.6%, and 74.2% of the provincial total emissions for Sb, As, and Se in 2014, respectively. The emissions from coal-fired power plants have been controlled by the installation of flue gas desulfurization systems. In addition, scenario analysis shows that Sb and As emissions from coal-fired power plants will decrease in the future in a high-efficiency control technology scenario. However, Se emissions in 2030 will still be higher than in 2014. This study demonstrates the importance of assessing the effectiveness of control measures and supplying necessary suggestions for managing coal combustion in Shandong.

**Keywords:** atmospheric emission; coal combustion; Sb, As, and Se; coal-fired power plants

## Introduction

Shandong Province is located in eastern China and has a very important function in the nation's development.

However, its developing industry and coal-dominated energy structure have induced numerous environmental problems, such as photochemical smog and haze [1]. Although Shandong's environmental statistics data show that air pollution was alleviated after enacting a series of pollution control policies, PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>2</sub>, and NO<sub>2</sub> are still major air pollutants. In 2014, PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>2</sub>, and

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NO<sub>2</sub> reached concentrations of 142, 82, 59, and 46 µg/m<sup>3</sup>, respectively [2] – higher than or close to the second-grade standard of the state (70, 35, 60, 40 µg/m<sup>3</sup> for PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>2</sub>, and NO<sub>2</sub>, respectively). Most air pollution comes from industry product processes where coal combustion is the key source [3, 4].

Trace elements (including Sb, As, and Se) are released from coal through smoke and are absorbed to particle matters in the atmosphere [5]. These trace elements will have a negative effect on human health via respiration. They will also travel long distances and be deposited into soils, water bodies, and plant leaves via wet and dry deposition, leading to a potential risk to the local environment [6-7]. The harmful impacts of Sb, As, and Se on the environment and public health were widely reported [8-11]. Cases of endemic arsenism caused by coal burning have been found in many Chinese provinces [6, 12], and more than 16,000 people have suffered from arsenism caused by coal burning [13]. Although Se is an essential trace element for humans and animals, high concentrations of it are harmful. Long-term exposure to high concentrations of Se may even result in hair and nail loss, and nervous system disorders [14]. Many cases of selenosis have been reported in regions with high Se concentrations [15]. Antimony and its compounds are listed as prior pollutants by the US Environmental Protection Agency and the EU because of its toxicity and suspected carcinogenic properties [10]. Antimony is released into the environment from coal combustion via two primary pathways: atmospheric emissions as volatile phases and the leachable portion of solid combustion by-products [16-17].

Coal combustion is one of the main anthropogenic sources of hazardous trace elements such as Sb, As, and Se [18-20], and their increased emissions with rising coal consumption has led to serious environmental pollution [21]. Given the large coal consumption in power plants and industry sectors in Shandong, Sb, As, and Se emissions are considered to be the highest in China [12, 14, 21].

Previous studies have focused primarily on conventional air pollutant emissions from coal combustion, such as SO<sub>2</sub>, NO<sub>x</sub>, PM, and Hg [20, 22-23]. Zhao et al. established a unit-based inventory of SO<sub>2</sub>, NO<sub>x</sub>, and PM emissions from coal-fired power plants in China [24]. Xiong et al. reported the emission inventories of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and Hg from coal-fired power plants in Shandong in 2012 [23]. Tian et al. found that Shandong is the biggest contributor to Sb, As, and Se emissions in China [13, 21, 25]. Meanwhile, the atmospheric emissions of Hg, As, and Se from coal combustion in China rapidly increased from 1980 to 2007, and Shandong is still the largest contributor [12, 16]. Cheng et al. [26] investigated the inventory of atmospheric emissions of Hg, As, Pb, Cd, and Cr from anthropogenic sources in China from 2000 to 2010, and found that coal consumption resulted in the ascending total emissions of heavy metals.

Various policies and regulations have been enacted in China to restrict the increasing emissions of trace elements from fuel combustion and industrial processes, including

the Emission Standard of Air Pollutants for Thermal Power Plants (GB13223-2011) [27-28] and the Air Pollution Control Plan for Shandong Province during 2013-20. Additionally, some specific targets were set with detailed technological methods to reduce the air pollutants in the province [23]. As a result, the adjustment and optimization of industrial structures occurred, consequently changing the emissions of Sb, As, and Se to some extent.

However, the emission inventories of Sb, As, and Se from coal combustion were evaluated at the national level. Few estimates of Sb, As, and Se emissions in Shandong Province have been published. In this paper evaluates the historical trends and characteristics of anthropogenic atmospheric emissions of Sb, As, and Se from coal combustion during 2005-14. Additionally, the temporal atmospheric emission inventories of Sb, As, and Se have been developed by economic sectors. Finally, the future emissions from coal-fired power plants are predicted by a scenario methodology.

## Methods and Materials

To evaluate the emissions of Sb, As, and Se from coal combustion, the emission amounts are calculated using different sectional coal consumption data and detailed emission factors. The basic equations can be expressed as follows:

$$EF_{i,a} = \sum_j \sum_m \sum_n C_{i,a} \times R_j \times (1 - P_{DC(m)}) \times (1 - P_{SO_2(n)}) \quad (1)$$

$$E_{i,a} = M_{i,a} \times EF_{i,a} \quad (2)$$

$$E_{a_{total}} = \sum_i E_{a,i} \quad (3)$$

... where  $EF_{i,a}$  is the emission factor of Sb, As, or Se from  $i$  sector in year  $a$ ;  $E_{i,a}$  is the emission of atmospheric Sb, As, or Se from  $i$  sector in year  $a$ ;  $E_{a_{total}}$  is the atmospheric emission of Sb, As, or Se in year  $a$ ;  $C$  is the average content of Sb, As, or Se in the coal consumed in Shandong;  $M$  is coal consumption;  $R$  is the fraction of Sb, As, or Se atmospherically released from coal combustion;  $P_{DC}$  and  $P_{SO_2}$  are the removal efficiencies of the installed dust collectors and desulfurization devices, respectively;  $i$  represents the emission source classified by economic sectors;  $j$  represents the boiler type;  $m$  represents the dust collector type;  $n$  represents the desulfurization devices type; and  $a$  is the year.

## Data Collection

The atmospheric emissions of Sb, As, and Se from coal combustion are the most important sources of anthropogenic discharge in Shandong [16, 29]. Coal is

Table 1. Release ratios of Sb, As, and Se from different boiler types.

Boiler type	Release rate (%)			References
	Sb	As	Se	
Pulverized-coal boiler	89.40	98.46	96.22	[9, 10, 26]
Stoker fired boiler	53.50	77.18	80.95	[9]
Fluidized-bed furnace	74.40	75.60	98.05	[9, 32, 38-39]
Coke furnace	70.00	30.00	40.00	[29, 40]

mainly consumed in four economic sectors, i.e., coal-fired power plants, industrial coal usage, residential coal usage, and other coal use. Therefore, coal consumption by those sectors was cited from the China Energy Statistical Yearbook and Shangdong Statistical Yearbook (2006-2015) [30-31].

### Category-Specific Emission Factors

The emission factors of Sb, As, and Se are determined by various factors, such as the patterns of combustion facilities and air pollution control devices (APCDs). Coal combustion facilities in the power plant, industrial, and other sectors can be divided into four types: stoker-fired boiler, circulating fluidized bed boiler, pulverized coal (PC) boiler, and coke furnace. The emission factors of Sb, As, and Se of each boiler type are listed in Table 1.

The APCDs include electrostatic precipitators (ESPs), fabric filters (FFs), cyclones, wet scrubbers, and flue gas desulfurization (FGD). Owing to the slight changes of PM/SO<sub>2</sub> removal efficiencies during the past decade [17, 18, 32-34], the constant average removal efficiencies of APCDs were adopted instead of the varied removal efficiencies. In fact, the most important factor that influences the removal of Sb, As, and Se emissions is the increased installation proportion of APCDs with a high PM/SO<sub>2</sub> removal efficiency. The removal efficiencies of Sb, As, and Se of different APCDs are presented in Table 2.

By the end of 2010, the proportion of ESPs installed in PC power units reached nearly 93%, while the FFs

Table 2. Removal efficiencies of Sb, As, and Se by different control devices.

Category	Removal efficiencies (%)			References
	Sb	As	Se	
ESP	83.5	86.20	73.78	[10]
FF	94.3	99.00	65	[10]
Cyclone	40	43.00	40	[10, 41]
WFGD	82.1	80.38	74.87	[10, 21]
Wet Scrubber	96.3	96.30	85	[10, 42]

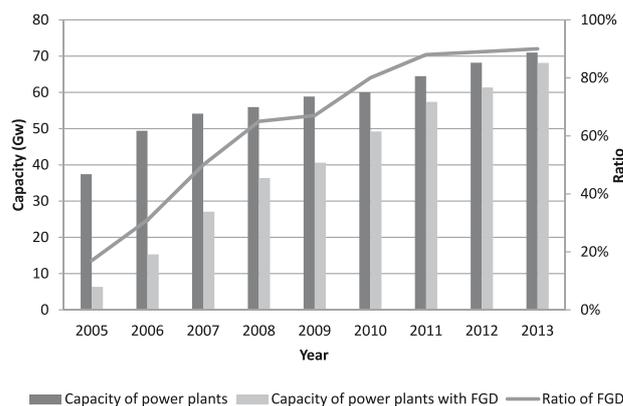


Fig. 1. The trend of proportion of power plants with FGD devices in Shandong, 2005-2014.

Data from Shandong Statistical Yearbook [30].

and electrical bag filters accounted for the remaining 7% [26]. All the fluidized bed furnaces were equipped with ESPs. The installation of FGD is very important to trace element removal [35]. Thus, the FGD installed ratio was considered in this paper, and data were collected from the China Electric Power Yearbook [31]. The increased proportion of power plants with FGD devices and the installed ratio in Shandong are shown in Fig. 1.

The average contents of Sb, As, and Se in consumptive coal are 0.71, 5.05, and 2.76 mg/kg, respectively, as calculated by Tian et al. [29, 36].

### Scenario Projections

To achieve the goal of energy conservation and emission reduction, three emission scenarios were developed for coal-fired power plants in Shandong to 2030 (Table 3) [1, 37]. All newly built units were assumed to be installed with wet FGD and nearly one-third equipped with FF/EF in scenarios [23]. In the business-as-usual scenario (BAU), emissions were evaluated by the current APCD installation ratios. The best-available-control-technology (BACT) is on the basis of the 2015 emission reduction target. The control technologies will continue to improve in the future. In the high-efficiency control technology (HECT), we assume that the control technology penetration would be stricter than that of the BACT scenario.

## Results and Discussion

### Coal Consumption

Coal consumption of the four economic sectors and the types of coal products are collected from the Shandong Energy Statistical Yearbooks from 2006 to 2015 [30]. In 2005, the total coal consumption in Shandong was only 260.6 Mt of raw coal, and 105.6 and 140.7 Mt by the coal-fired power plants and industrial sectors, respectively. Because of the relatively rapid economic growth in

Table 3. Assumptions of activity levels and control technology penetrations.

Years	Scenarios	Coal consumption (Mt)	WFGD	ESP	FF/EF
2014	BASE	146.57	91%	99%	1%
2017	BAU	186.32	93%	92%	8%
	BACT	186.32	94%	91%	9%
	HECT	186.32	96%	90%	10%
2020	BAU	232.3	94%	86%	14%
	BACT	232.3	96%	81%	19%
	HECT	232.3	99%	77%	23%
2025	BAU	259.7	95%	82%	18%
	BACT	259.7	99%	70%	30%
	HECT	259.7	100%	65%	35%
2030	BAU	267.5	93%	80%	20%
	BACT	267.5	100%	60%	40%
	HECT	267.5	100%	56%	44%

Shandong, its coal consumption increased steadily and quickly, especially from 2005 to 2010 (Fig. 2). By the end of 2014, the total coal consumption in Shandong reached 395.6 Mt. Moreover, the power plant sector was the largest consumer in total coal consumption, followed by the industrial sector. Residential use and other use sectors accounted for only a small proportion of the total coal consumption in Shandong. However, the growth rate has been falling since 2010, and total coal consumption decreased after 2011. The major reason for the decrease is the change in energy consumption composition.

In China, although total energy consumption increased rapidly after 2001, the percentage of coal consumption declined after 2006, and to 66% in 2013 (Fig. 3). Both the percentages of natural gas and green energy (such as hydropower, nuclear power, and wind power) increased

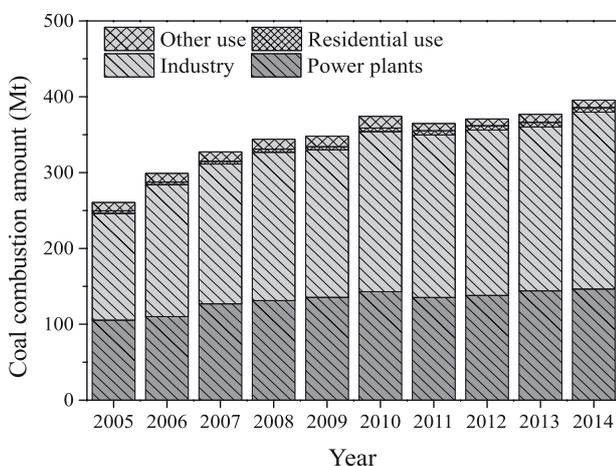


Fig. 2. Trend of coal consumption by different sectors in Shandong province, 2005-2014.

during the mentioned years. Specifically, the percentage of green energy continued to increase to 11.2% by 2014, with an annual growth rate of 5.00%. Thus the use of clean and high-quality energy, such as hydro-power, natural gas, wind power, and nuclear power, was introduced to optimize the energy structure.

#### Emission Factors of Sb, As, and Se from Coal-fired Power Plants

The emission factors of Sb, As, and Se from coal-fired power plants are related to its concentrations in coals, coal-burning modes, and types and quantities of installed APCDs. The final combined emission factors of Sb, As, and Se are calculated to reflect the emission levels through burning per Mt coal in power plants in Shandong (Fig. 4). The larger the final combined emission factors, the larger the amount of emissions from the burning of every ton of raw coal. Emission factors are directly proportional to the Sb, As, and Se contents in coal and the release rates, but inversely related to the removal efficiencies of APCDs. The emission factors of these three elements declined with

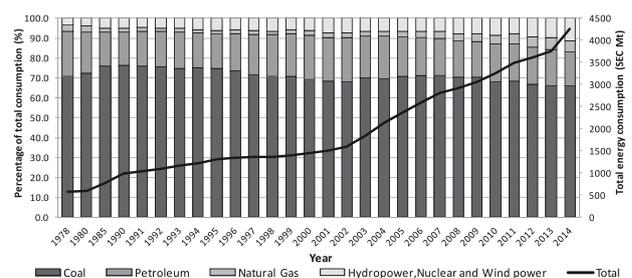


Fig. 3. Trend of total consumption of energy and its composition in China, 1978-2014.

Data from China Statistical Yearbook 2015 [31].

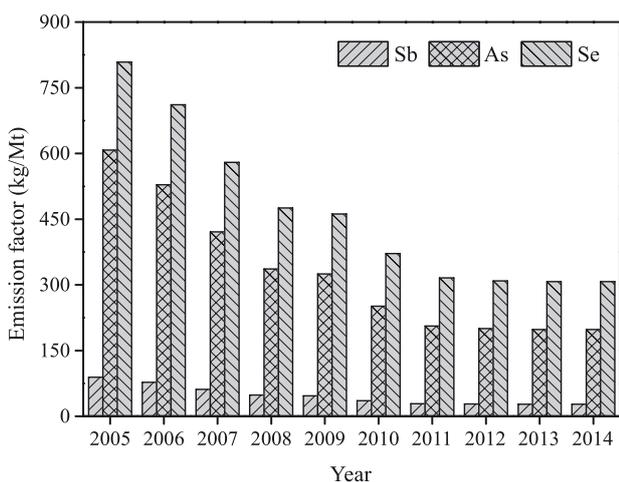


Fig. 4. Emission factors of Sb, As, and Se from coal-fired power plants of Shandong, 2005-2014.

the installed FGD equipment every year. A remarkable difference exists not only in the emission factors of different years, but also among Sb, As, and Se.

#### Emissions by Sectors

The trends of Sb, As, and Se emissions by different economic sectors from 2005 to 2014 are illustrated in Figs 3-5, respectively. The provincial total emissions of Sb, As, and Se from coal in 2005 are estimated at 40.26, 246.5, and 255.9 t, and increased to 51.35, 311.9, and 313.9 t in 2014, with annual growth rates of 2.75%, 2.65%, and 2.27%, respectively.

The Sb emissions from the four economic sectors was calculated (Fig. 5). The power plants and industrial use are two major contributors to atmospheric Sb emissions, especially for industrial use. However, Sb emissions from the power plant sector have decreased every year. For example, Sb emissions from coal-fired power plants totaled 9.45 t in 2005, contributing to 23.5% of total Sb

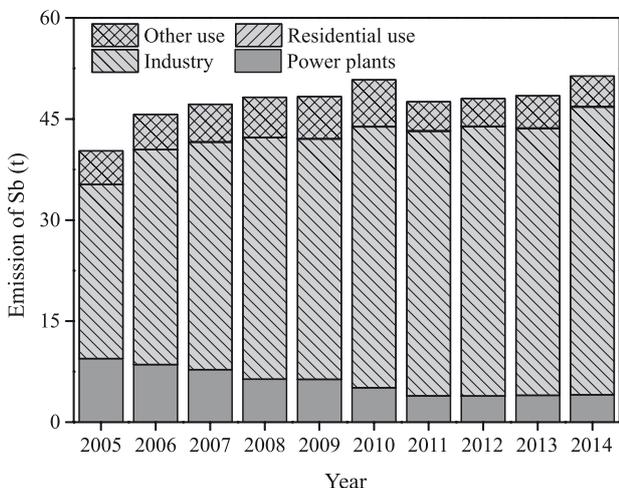


Fig. 5. Historical trends of annual Sb emissions, 2005-2014.

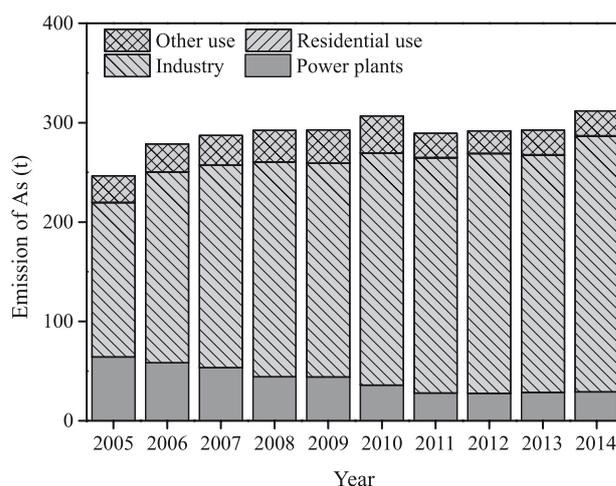


Fig. 6. Historical trends of annual As emissions, 2005-2014.

emissions. And Sb emissions decreased to 4.05 t in 2014, accounting for only 7.89 % of the total. However, Sb emissions in industrial use reached 42.73 t in 2014, with an average annual growth rate of 7.28%.

The emissions of As and Se from the four economic sectors are shown in Figs 6, 7. The trends of As and Se emissions from the power plant sector are similar to that of Sb. The emissions of As and Se reached 64.16 and 85.35 t, contributing to 26.0% and 33.3% of total emissions in 2005. And its amounts decreased to 29.04 and 45.06 t in 2014. However, the emissions of As and Se from the industrial use sector have been increasing since 2005, reaching 257.6 and 233.1 t in 2014. In addition, residential use sector emissions of As and Se increased from 2005 to 2014. Because of the use of clean fuels such as natural gas and electricity, especially in urban areas, other uses of coal decreased after 2010. Accordingly, the emissions of Sb, As, and Se dropped. The emissions of Sb, As, and Se are estimated to decrease with coal consumption reductions in the other use sector, including farming, construction, transportation, and commerce. The emissions of Sb, As,

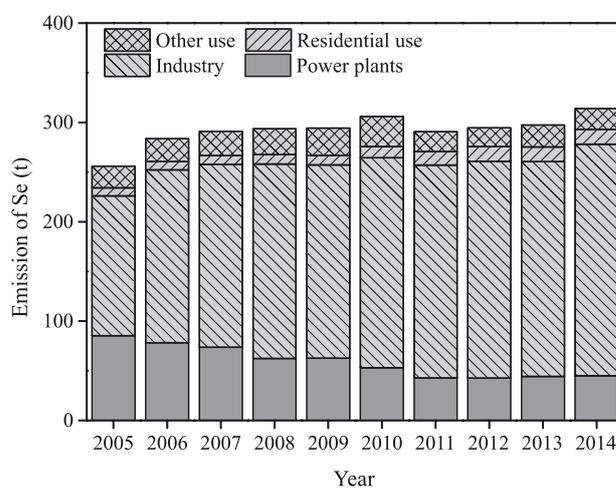


Fig. 7. Historical trends of annual Se emissions, 2005-2014.

and Se from other uses reached 4.51, 25.0, and 20.9 t in 2014, respectively.

The reasons for the results are:

1. Power plants and industrial use sectors are the main consumers in Shandong.
2. The decreased emissions of Sb, As, and Se from the power plant sector are ascribed to the change in emission factors as a result of the increasing number of power plants installed with APCDs, including ESPs and the FGD system.
3. The industrial use sector contains a significantly higher share of uncontrolled or poorly controlled boilers.
4. The rapid development of construction, transportation, and commerce enhances the emissions of Sb, As, and Se from other sectors.

With the start of the 11<sup>th</sup> Five Years Plan (FYP) (2006-2010), the atmospheric emissions of heavy metals in China have grown at a more moderate pace owing to the comprehensive effects of the increasing application of advanced APCDs and the slower growth of fuel consumption and output of industrial products [26]. From 2001 to 2005, the annual increasing rate of coal consumption in Shandong was 26.9%, but only 6.27% during 2006 and 2010. Moreover, the change in energy structure – especially the increase in clean energy – constrained the growth of coal consumption. The industrial use sector became the largest consumer with the highest annual growth rate (Fig. 2). The electricity output from clean energy in Shandong, including hydropower, nuclear power, and wind power, is rapidly developing (Fig. 8).

#### Scenario Analysis for Future Emissions from Coal-Fired Power Plants

Air pollution policies, especially for controlling air pollutant emission from coal-fired power plants, were issued during the 12<sup>th</sup> FYP [1, 27-28]. Therefore, the emissions of Sb, As, and Se from coal-fired power plants could be alleviated with the tightening controls in Shandong. Based on the three scenarios, the predicted emissions from the coal power sector under these three

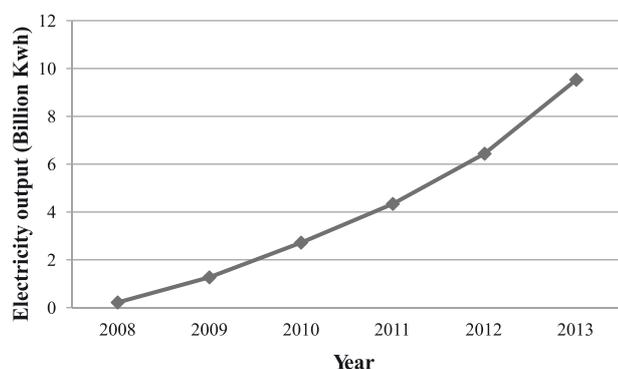


Fig. 8. Trend of electricity output from clean energy (hydropower, nuclear power, wind power, and others) in Shandong, 2008-2013. Data from Shandong Statistical Yearbook [30].

Table 4. Projected emissions (tons) from Shandong coal-fired power plants in 2017, 2020, 2025, and 2030 under three scenarios.

Years	Scenarios	Sb	As	Se
2014	BASE	3.86	27.72	43.27
2017	BAU	4.37	30.952	53.31
	BACT	4.19	29.66	52.05
	HECT	3.86	27.43	49.45
2020	BAU	5.04	35.10	65.18
	BACT	4.51	31.05	61.66
	HECT	3.89	26.52	56.01
2025	BAU	5.28	36.34	70.94
	BACT	4.09	27.20	61.13
	HECT	3.75	24.45	57.47
2030	BAU	5.76	39.08	76.95
	BACT	3.70	23.45	57.12
	HECT	3.57	22.07	55.19

types of control levels are still high (Table 4, Figs. 9-10). The emissions of Sb, As, and Se increase by 13.4%, 11.6%, and 23.2% in the 2017 BAU scenario compared with that of the 2014 BASE scenario, with a 27.6% increase in coal consumption. The emissions of Sb, As, and Se from coal-fired power plants in the BACT scenario increase by 8.69%, 7.03%, and 20.3% compared to the baseline emissions in 2014.

However, in the HECT scenario of 2017, emissions of As were reduced by 1.01%, while Se emissions increased by 14.3%. In contrast, the emissions of Sb and As from coal-fired power plants will decrease in the 2030 BACT and 2030 HECT scenarios, with 40% and 44% ratios of installed FFs, respectively. Due to the relatively lower co-benefit removal efficiency of ESP/FFs and WFGD (Table 2), the Se emissions under the BACT and HECT

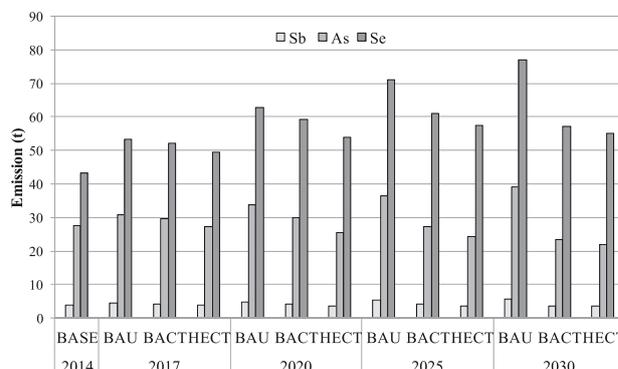


Fig. 9. Projected Sb, As, and Se emissions from coal-fired power plants in Shandong Province in 2017, 2020, 2025, and 2030 under three scenarios.

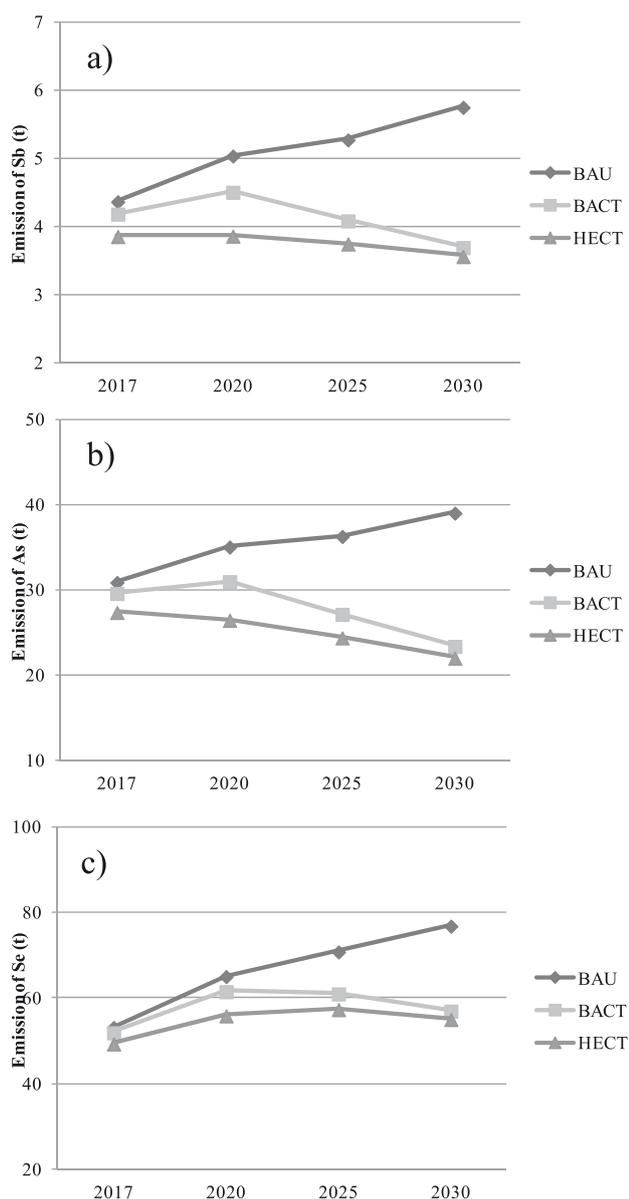


Fig. 10. Projected Sb (a), As (b), and Se (c) emissions from Shandong coal-fired power plants.

scenarios in 2030 are still higher than those in 2014. Fortunately, the predicted Se emissions under the HECT scenarios will begin to decrease after 2025. Thus, the FFs combined with the WFGD system is the best choice to control the emissions of Sb, As, and Se to date. In the future, innovative multi-pollutant control technology may be available as the more cost-effective way to further diminish trace element emissions.

#### Uncertainties Analysis

The emissions of Sb, As, and Se are affected by several factors, and some uncertainties remain in the estimation of Sb, As, and Se emissions from coal combustion in Shandong. Firstly, the significant discrepancies are

observed in the estimates of the typical Sb, As, and Se contents in coals. Secondly, the combustion release rates and emission factors at different burners are improvable. Finally, the determination of the removal rates of Sb, As, and Se through the existing APCDs, such as ESP/FFs and FGD, is difficult. The removal rates of Sb, As, and Se by various control devices, such as WFGD, highly depend on the speciations of these elements in flue gas (elemental vs. oxidized forms) [18]. Oxidized metals can be easily removed in WFGD compared with its elemental forms. Coal properties, such as Cl content, affect the element speciation in the flue gas and in turn, the removal rate in WFGD [33]. Currently, actual measurements of Sb, As, and Se emission rates and species profiles from Shandong combustion facilities are still very limited, and the capture performance of Sb, As, and Se in Chinese emission control devices as well, especially for As and Se. The remaining uncertainties of our inventories can be reduced in future by additional field testing data for all types of coal and combustion facilities.

#### Conclusions

The calculated provincial total atmospheric emissions of Sb, As, and Se from coal combustion in Shandong rapidly increased from 40.26, 246.5, and 255.9 t in 2005 to 51.35, 311.9, and 313.9 t in 2014, with annual growth rates of 2.75%, 2.65%, and 2.27%, respectively. Industrial use is the largest single contributor. Emissions of Sb, As, and Se from this sector reached 42.73, 257.6, and 233.1 t in 2014, accounting for 83.2%, 82.6%, and 74.2% of the total emissions, respectively. The power plant sector is the second sector, contributing 23.5%, 26.0%, and 33.3% of the total emissions of Sb, As, and Se in 2005, respectively. However, these amounts decreased to 7.89%, 9.31%, and 14.3% in 2014.

Compared to the 2014 baseline, Sb and As would begin to decline from 2020 under the BACT and HECT scenarios, but emissions of Se in 2025 will still be higher than in 2014 in Shandong. By 2030, the approximate emission reductions of 7.43% for Sb and 20.4% for As can be achieved under the HECT scenario. Thus, the emissions of Sb, As, and Se from coal-fired power plants can be controlled with the development of desulfurization and dust-removing devices.

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## References

- The People's Government of Shandong Province (PGSD) The Air Pollution Control Plan for Shandong Province during 2013-2020 (in Chinese), **2013**. [http://sdgb.shandong.gov.cn/art/2013/9/3/art\\_4563\\_1906.html](http://sdgb.shandong.gov.cn/art/2013/9/3/art_4563_1906.html).
- Shandong Province Environmental Bulletin, Environmental Protection Department of Shandong Province, **2014**. <http://xxgk.sdein.gov.cn/xxgkml/hjzkgb/201506/t20150602.html>.
- Shandong environment statistical data, Environmental protection department of Shandong province, **2014**. [http://zlc.sdein.gov.cn/hjtj/201511/t20151103\\_286781.html](http://zlc.sdein.gov.cn/hjtj/201511/t20151103_286781.html).
- CHENG H., ZHOU T., LI Q., LU L., LIN C. Anthropogenic Chromium Emissions in China from 1990 to 2009. *PLoS ONE* **9**: e87753, **2014**.
- LI H., WANG Q.G., SHAO M., WANG J., WANG C. Fractionation of airborne particulate-bound elements in haze-fog episode and associated health risks in a megacity of southeast China. *Environmental Pollution* **208**, Part B: 655, **2016**.
- LI H., WANG J., WANG Q.G., QIAN X., QIAN Y. Chemical fractionation of arsenic and heavy metals in fine particle matter and its implications for risk assessment: A case study in Nanjing, China. *Atmospheric Environment* **103**, 339, **2015**.
- LI H., QIAN X., WANG Q. Heavy Metals in Atmospheric Particulate Matter: A Comprehensive Understanding Is Needed for Monitoring and Risk Mitigation. *Environmental Science & Technology* **47**, 13210, **2013**.
- FILELLA M., BELZILE N., CHEN Y.W. Antimony in the environment: a review focused on natural waters I. Occurrence. *Earth-Science Reviews* **57**, 125, **2002**.
- FILELLA M., BELZILE N., LETT M.C. Antimony in the environment: A review focused on natural waters. III. Microbiota relevant interactions. *Earth-Science Reviews* **80**, 195, **2007**.
- GEBEL T. Arsenic and antimony: comparative approach on mechanistic toxicology. *Chemico-Biological Interactions* **107**, 131, **1997**.
- RAYMAN M.P. Selenium and human health. *Lancet* **379**: 1256, **2012**.
- TIAN H.Z., WANG Y., XUE Z.G., CHENG K., QU Y.P. Trend and characteristics of atmospheric emissions of Hg, As, and Se from coal combustion in China, 1980-2007. *Atmos Chem Phys* **10**, 11905, **2010**.
- National Health and Family Planning statistical yearbook, National Health and Family Planning Commission of the People's Republic of China, **2014**, <http://www.nhfpc.gov.cn/zwgkzt/tjnj/list.shtml>.
- ZHU J., WANG N., LI S., LI L., SU H. Distribution and transport of selenium in Yutangba, China: Impact of human activities. *Science of The Total Environment* **392**, 252, **2008**.
- MAO D., SU H., YAN L. An epidemiologic investigation on selenium poisoning in southwestern Hubei Province. *Chinese Journal of Endemiology* **9** (5), 311, **1990**.
- TIAN H.Z., ZHAO D., HE M.C., WANG Y., CHENG K. Temporal and spatial distribution of atmospheric antimony emission inventories from coal combustion in China. *Environmental Pollution* **159**, 1613, **2011**.
- MEIJ R., WINKEL H. The emissions of heavy metals and persistent organic pollutants from modern coal-fired power stations. *Atmospheric Environment* **41**, 9262, **2007**.
- ONDOV J.M., RAGAINI R.C., BIERMANN A.H. Emissions and particle-size distributions of minor and trace elements at two western coal-fired power plants equipped with cold-side electrostatic precipitators. *Environmental Science & Technology* **14**, 1534, **1980**.
- SENIOR C.L., HELBLE J.J., SAROFIM A.F. Emissions of mercury, trace elements, and fine particles from stationary combustion sources. *Fuel Processing Technology* **65-66**, 263, **2000**.
- YOKOYAMA T., ASAKURA K., MATSUDA H., ITO S., NODA N. Mercury emissions from a coal-fired power plant in Japan. *Science of The Total Environment* **259**, 97, **2000**.
- CHEN J., LIU G., KANG Y., WU B., SUN R., ZHOU C., WU D. Atmospheric emissions of F, As, Se, Hg, and Sb from coal-fired power and heat generation in China, *Chemosphere*, **90**, 1925, **2013**.
- TANG S., FENG X., QIU J., YIN G., YANG Z. Mercury speciation and emissions from coal combustion in Guiyang, southwest China. *Environmental Research* **105**, 175, **2007**.
- XIONG T., JIANG W., GAO W. Current status and prediction of major atmospheric emissions from coal-fired power plants in Shandong Province, China. *Atmospheric Environment* **124**, Part A: 46, **2016**.
- ZHAO Y., WANG S., DUAN L., LEI Y., CAO P. Primary air pollutant emissions of coal-fired power plants in China: Current status and future prediction. *Atmospheric Environment* **42**, 8442, **2008**.
- TIAN H., QU Y., WANG Y., PAN D., WANG X. Atmospheric selenium emission inventories from coal combustion in China in 2005. *China Environmental Science* **29**, 1011, **2009**.
- CHENG K., WANG Y., TIAN H., GAO X., ZHANG Y. Atmospheric Emission Characteristics and Control Policies of Five Precedent-Controlled Toxic Heavy Metals from Anthropogenic Sources in China. *Environmental Science & Technology* **49**, 1206, **2015**.
- Ministry of Environmental Protection of China (MEP) Emission Standard of Air Pollutants for Thermal Power Plants. GB 13223-2011, Beijing, China, **2011**.
- Ministry of Environmental Protection of China (MEP) Ambient Air Quality Standards; GB 3095-2012. Beijing, China, **2012**.
- TIAN H.Z., ZHU C.Y., GAO J.J., CHENG K., HAO J.M. Quantitative assessment of atmospheric emissions of toxic heavy metals from anthropogenic sources in China: historical trend, spatial distribution, uncertainties, and control policies. *Atmos Chem Phys* **15**, 10127, **2015**.
- Shandong Statistics Bureau (SDSB), Shandong Statistical Yearbook 2006-2015 (in Chinese). Beijing: China Statistics Press, **2006-2015**.
- China Electricity Council (CEC), China electric power yearbook. Beijing: China Electric Power Press, **2005-2015**.
- HELBLE J.J., MOJTAHEDI W., LYRÄNEN J., JOKINIEMI J., KAUPPINEN E. Trace element partitioning during coal gasification. *Fuel* **75**, 931, **1996**.
- KLIKA Z., BARTOŇOVÁ L., SPEARS D.A. Effect of boiler output on trace element partitioning during coal combustion in two fluidised-bed power stations. *Fuel* **80**, 907, **2001**.
- GOGEBAKAN Z., GOGEBAKAN Y., SELÇUK N., SELÇUK E. Investigation of ash deposition in a pilot-scale fluidized bed combustor co-firing biomass with lignite. *Bioresource Technology* **100**, 1033, **2009**.
- OTERO-REY J.R., LÓPEZ-VILARIÑO J.M., MOREDA-PIÑEIRO J., ALONSO-RODRÍGUEZ E., MUNIATEGUI-LORENZO S. As, Hg, and Se Flue Gas Sampling in a Coal-Fired Power Plant and Their Fate during Coal Combustion. *Environmental Science & Technology* **37**, 5262, **2003**.
- DAI S., REN D., CHOU C., FINKELMAN R., SEREDIN V., ZHOU Y. Geochemistry of trace elements in Chinese coals A review of abundances, genetic types, impacts on

- human health, and industrial utilization, *Int. J. Coal Geol.* **94**, 3, **2012**.
37. Shandong Statistics Bureau (SDSB), Shandong Statistical Yearbook 2013. Beijing: China Statistics Press, **2013**.
38. DEMIR I., HUGHES R.E., DEMARIS P.J. Formation and use of coal combustion residues from three types of power plants burning Illinois coals. *Fuel* **80**, 1659, **2001**.
39. AMAND L.E., LECKNER B. Metal emissions from co-combustion of sewage sludge and coal/wood in fluidized bed. *Fuel* **83**, 1803, **2004**.
40. ZAJUSZ-ZUBEK E., KONIECZYŃSKI J. Dynamics of trace elements release in a coal pyrolysis process. *Fuel* **82**, 1281, **2003**.
41. GOGEBAKAN Z., SELÇUK N. Trace elements partitioning during co-firing biomass with lignite in a pilot-scale fluidized bed combustor. *Journal of Hazardous Materials* **162**, 1129, **2009**.
42. ONDOV J.M., RAGAINI R.C., BIERMANN A.H. Elemental emissions from a coal-fired power plant. Comparison of a venturi wet scrubber system with a cold-side electrostatic precipitator. *Environmental Science & Technology* **13**, 598, **1979**.