HAL
open science

Long-Term Trends of Anthropogenic SO2 , NOx , CO, and NMVOCs Emissions in China

W. Sun, M. Shao, Claire Granier, Y. Liu, C. Ye, Y. Zheng

To cite this version:

W. Sun, M. Shao, Claire Granier, Y. Liu, C. Ye, et al.. Long-Term Trends of Anthropogenic SO2 , NOx , CO, and NMVOCs Emissions in China. Earth's Future, 2018, 6 (8), pp.1112-1133. 10.1029/2018EF000822 . hal-02115996

HAL Id: hal-02115996

https://hal.science/hal-02115996v1

Submitted on 16 Jun 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Copyright

Check for updates

AGU100 ADVANCING EARTH AND SPACE SCIENCE

Earth's Future

RESEARCH ARTICLE
10.1029/2018EF000822

Key Points:
- Historical emission trends were evaluated for SO_2, NO_x, CO, and volatile organic compounds (VOCs) in China since 1949
- National SO_2 and NO_x emissions started to decline in 2007 and 2012, respectively, while VOCs emissions kept on rising
- A reactivity-based control strategy was proposed to achieve greater OFP reduction with less VOCs emissions reduction

Supporting Information:
- Supporting Information S1
- Table S1

Correspondence to:
M. Shao,
mshao@pku.edu.cn

Citation:
Sun, W., Shao, M., Granier, C., Liu, Y., Ye, C. S., & Zheng, J. Y. (2018). Long-term trends of anthropogenic SO_2, NO_x, CO, and NMVOCs emissions in China. *Earth's Future*, *6*, 1112–1133. https://doi.org/10.1029/2018EF000822

Received 21 JAN 2018
Accepted 24 JUN 2018
Accepted article online 29 JUN 2018
Published online 21 AUG 2018

©2018. The Authors.
This is an open access article under the terms of the Creative Commons Attribution-NonCommercial-NoDerivs License, which permits use and distribution in any medium, provided the original work is properly cited, the use is non-commercial and no modifications or adaptations are made.

Long-Term Trends of Anthropogenic SO_2, NO_x, CO, and NMVOCs Emissions in China

W. Sun[1], M. Shao[2,1], C. Granier[3,4,5], Y. Liu[1], C. S. Ye[1], and J. Y. Zheng[2]

[1]State Joint Key Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing, China, [2]Institute for Environmental and Climate Research, Jinan University, Guangzhou, China, [3]Laboratoire d'Aérologie, University of Toulouse, CNRS, UPS, Toulouse, France, [4]Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, USA, [5]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, CO, USA

Abstract Studies reporting the historical trends of SO_2, NO_x, CO, and nonmethane volatile organic compounds emissions in China using unified approaches and sources are limited. Here we established 66-year emission trends of these four species in China. Six primary anthropogenic sources were included, and we made a series of improvements to the few existing inventories based on detailed statistical data, recently published emission factors, and technology renewal to reduce the uncertainties. National SO_2, NO_x, CO, and nonmethane volatile organic compounds emissions in 2015 were 27.1, 20.6, 188, and 28.4 Mt, with annual growth rates of 5.8%, 5.9%, 3.8%, and 4.6% since 1949, respectively. In recent years, fossil fuel combustion was the major contributor to SO_2, NO_x, and CO emissions, whereas industrial process contributed most to VOCs emissions. Our results revealed a 10% decrease in the SO_2 emissions from 2005 to 2010 as a result of improvements in the flue gas desulfurization installation rate. NO_x emissions stopped rising and started falling in 2011, with technology renewal and the penetration of end-of-pipe control measures in industrial boilers and cement production. Furthermore, we calculated future speciated VOCs emissions and ozone formation potential under alternative policy scenarios and projected to reduce emissions by 10% (compared with 2013 levels) with stringent control measures in 2020. A reactivity-based control strategy was proposed to achieve greater ozone formation potential reductions while requiring less VOCs emissions reduction.

Plain Language Summary Over past half century, the emissions of air pollutants generated through human activities in China have increased rapidly. The study of long-term emissions trends will improve our understanding of human influence on climate change and air quality. Here we established 66-year emission trends of four major species for the first time and evaluated the impact of government policy on emission. The central government initialed the most stringent ever action plan in China to cut emissions, for example, from 1 January 2018, China began to levy environmental protection tax, aimed at SO_2, NO_x, CO, VOCs, and other pollutants. Our result reveals a decrease in the SO_2 and NO_x emissions, consistent with satellite observation, while VOCs emissions continued to rise. The emission-based reduction plan for VOCs have been implemented nationwide. In this study, we further proposed a reactivity-based control strategy to improve the air quality more effectively.

1. Introduction

In 2013, the radiative forcing of anthropogenic compounds was reported by the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report. Emissions of short-lived gases, including SO_2, NO_x, CO, and nonmethane volatile organic compounds (NMVOCs), contribute significantly to the total anthropogenic radiative forcing (IPCC, 2013). Developing countries, such as China, India, and Russia, are the largest emitters of these pollutants and usually lack data on historical emissions trends (Akimoto, 2003; Stamenkovic et al., 2016). In October 2013 the panel set up a task group to help the IPCC continue to improve its operation and products and one of its goals was to develop options and ways to enhance the participation and contribution of developing countries in the future work of the IPCC. Therefore, the purposes of our study is to track historical emissions trends of SO_2, NO_x, CO, and VOCs in China.

In order to study regional air pollution problems, researchers have developed several bottom-up emissions inventories in China. Jernelov (1983) estimated the total SO_2 emissions in China to be about 15 million metric tons (Mt) per year in a study on acid rain. Kato and Akimoto (1992) calculated the anthropogenic emissions of

SO_2 and NO_x in East Asia in 1975, 1980, 1985, 1986, and 1987 and found that China was the largest emitter of those two pollutants in 1987. Streets and Waldhoff (2000) estimated the emissions of three of the major air pollutants in China—SO_2, NO_x, and CO—for the years 1990, 1995, and 2020 under different assumptions about the level of environmental controls. To quantify the chemical transport of Asian pollution and assess its implications for regional air quality, the Transport and Chemical Evolution over the Pacific experiment (Carmichael et al., 2003) and the Intercontinental Chemical Transport Experiment-Phase B mission (Q. Zhang et al., 2009) compiled East Asian emissions inventories of SO_2, NO_x, VOCs, fine particulate matter ($PM_{2.5}$), black carbon, and CO in 2000, 2001, and 2006. Along with increasingly serious local photochemical smog pollution, multiyear emissions inventories of VOCs in China for 1980–2005 and 2008–2012 were established (Bo et al., 2008; Wu et al., 2016), and speciated VOCs emissions were calculated in 2013, using source profiles from the U.S. SPECIATE database. Tsinghua University developed the Multi-resolution Emission Inventory for China (http://www.meicmodel.org) to calculate air pollutants and CO_2 emissions in China, including speciated VOCs, based on locally measured profiles and SPECIATE data (Li, Zhang, et al., 2017; Li, Zhang, et al., 2014).

Detailed studies on the emissions inventories of SO_2, NO_x, CO, and VOCs have been published; however, there are significant differences among the results. Most inventories focused on emissions in China for a single year or over a few years and calculated speciated VOCs based on source profiles from developed countries (Li, Zhang, et al., 2014; Wu & Xie, 2017). Therefore, our goal is to quantify historical SO_2, CO, NO_x, and VOCs emissions trends from 1949 to 2015 in China using a uniform approach and sources. In this study, we selected a technology-based methodology for different pollutants and sources. Emissions factors (EFs) were updated with local measurements and model calculations, and most activities were drawn from first-hand statistical data to reduce uncertainty. Seventy percent of the VOC source profiles were from local measurements (Mo et al., 2016). The historical inventory for the earlier years of the study period was estimated based on time-varying statistical data and the technological level available at the time. Additionally, many new sources were added, including nonroad mobile sources, sewage treatment, and agrochemical use. Finally, we summarized several major inventories and official statistical results and compared them with the results of our study.

Widespread and persistent ozone pollution has been one of the most serious types of pollution in eastern China owing to increasing emissions of O_3 precursors (mainly VOCs and NO_x; Li, Bei, et al., 2017; Wang et al., 2017; Wu & Xie, 2017; Xue et al., 2014). The State Council of China issued an Air Pollution Prevention and Control Action Plan, which included SO_2, NO_x, and VOCs into the targets of emissions control in 2013. In 2016, the goal was to reduce SO_2 emissions by 15%, NO_x emissions by 15%, and VOCs emissions by 10% during the period of the 13th Five-Year Plan (FYP, 2016–2020). A major industrial VOCs reduction plan was issued by the Ministry of Industry and Information Technology (MIIT) and the Ministry of Finance in July 2016, hereafter called the MIIT Plan, which calls for a reduction of 3.3 Mt of VOCs from 2016 to 2018. Many studies have shown that reactivity-based VOCs control measures are more efficient in air quality improvement (Mo et al., 2016; Ou et al., 2015; Wu & Xie, 2017). Therefore, in this study, we evaluated the effects of the VOCs emissions reduction strategies outlined in the MIIT Plan and proposed a reactivity-based control scenario with greater reduction of ozone formation potential (OFP) while requiring less VOCs emissions reduction.

2. Methods and Data

Many methods have been used to establish anthropogenic emissions inventories, including the material balance method, EFs, forward modeling, and inverse modeling, which are applicable to different pollutants and different spatial scales. The material balance method, which is based on the principle of mass conservation, is suitable for pollutants with a single component and a simple and clear process, such as SO_2, NOx, and some greenhouse gases, combined with emissions sources, industrial production and management, recycling, and environmental controls, to comprehensively estimate the generation and emission of pollutants in the production process. More than 90% of China's SO_2 emissions come from the combustion of coal, oil, and other fuels (Su et al., 2011), from simple generation and management processes. Thus, we chose the material balance method to calculate SO_2 emissions from fossil fuel combustion, using EFs to calculate CO, NO_x, and VOCs emissions from 1949 to 2013 in national level and proposed a reactivity-based control strategy

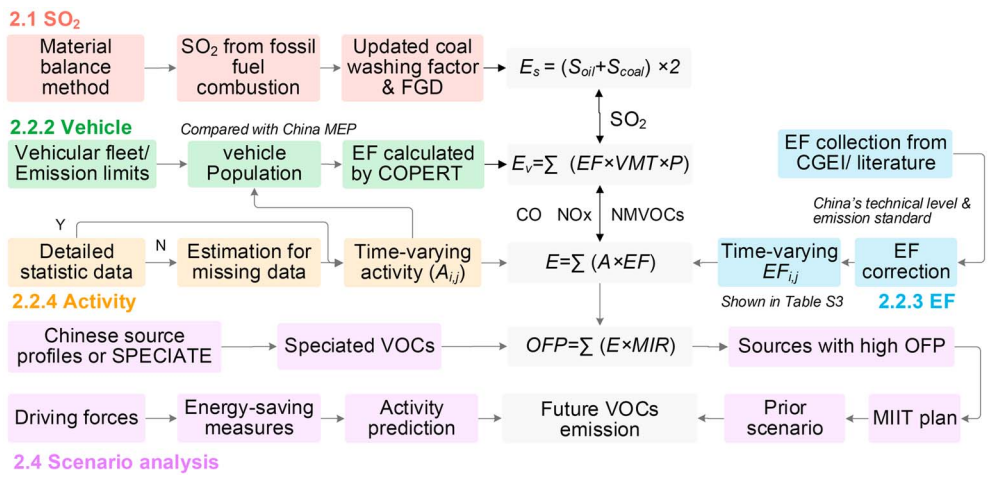

Figure 1. Schematic of emissions calculations used in this study. Red boxes indicate SO_2 emissions calculated by the material balance method. Green boxes indicate calculations of vehicular emissions. Yellow and blue boxes indicate simplified estimations of activities and EFs, respectively. Purple boxes indicate estimations of speciated VOCs emissions and OFPs, and scenario analysis. EF = emission factor; FGD = flue gas desulfurization; CGEI = Compilation Guide for the Air Pollutant Emission Inventory; MIIT = Ministry of Industry and Information Technology; MIR = maximum incremental reactivity; NMVOC = nonmethane volatile organic compound; OFP = ozone formation potential; VMT = vehicle mileage traveled.

based on speciated VOCs emissions and OFPs. Figure 1 summarizes the method adopted in this study. This Chinese emissions inventory covers 31 provinces, excluding Taiwan, Hong Kong, and Macao.

2.1. Mass Balance Method

The primary SO_2 emissions sources were vehicles exhaust, coal, and oil combustion. Vehicular emissions accounted for less than 5% and calculated by equation (1) with COPERT4 model, which will be introduced in section 2.2.2. The material balance model was used to estimate the emissions of fossil fuel combustion (see Text S1 in the supporting information for detailed estimation and formula), based on coal and oil consumption, sulfur content, combustion residue, coal washing factors, and flue gas desulfurization (FGD) removal efficiency. We made some improvements and revisions to the previous mathematical model and updated the database to reflect rapid technology renewal and new environmental standards.

We employed three improved measures for establishing a new SO_2 inventory for coal and oil combustion in China. First, we reexamined the energy statistics and corrected for coal and oil consumption instead of production, based on data from the China Energy Statistical Yearbook (CESYs; National Bureau of Statistics, 1986, 1989, 1991–2016). Second, we revised the sulfur content of heavy oil based on national standards SY1091-77 (88) and SH/T0356-1996, to 2% in 1949, 1% in 1996, and 0.5% in 2013. We estimated the sulfur content of coal as 1.35% before 1985, 1.27% in 1990, 1.12% in 1995, 1.08 in 2000, 1.02% in 2005, and 0.998% in 2010, based on the data reported in literatures (Klimont et al., 2009; Kurokawa et al., 2013; Lu et al., 2010; Su et al., 2011) and the promotion of low-sulfur coal policy. Finally, we updated the FGD loading ratio and removal efficiency from the China Electric Power Yearbook (China Electricity Council, 1993–2016), instead of interpolating estimations, to reduce the uncertainty of the results, because the promotion of FGD devices is an important factor in reducing SO_2 emissions.

2.2. EF Method

2.2.1. General Methodology

Anthropogenic emissions were divided into a series of sources and sectors according to pollutant categories, economic sectors, and technical characteristics (see Table S1 in the supporting information for the full list of classification). VOCs emissions were classified into six sources: vehicles, fossil fuel combustion, industrial processes, biomass burning, solvent use, and nonroad mobile sources. NO_x emissions were classified into four sources: vehicles, fossil fuel combustion, biomass burning, and nonroad mobile sources. CO emissions were classified into five sources: vehicles, fossil fuel combustion, industrial processes,

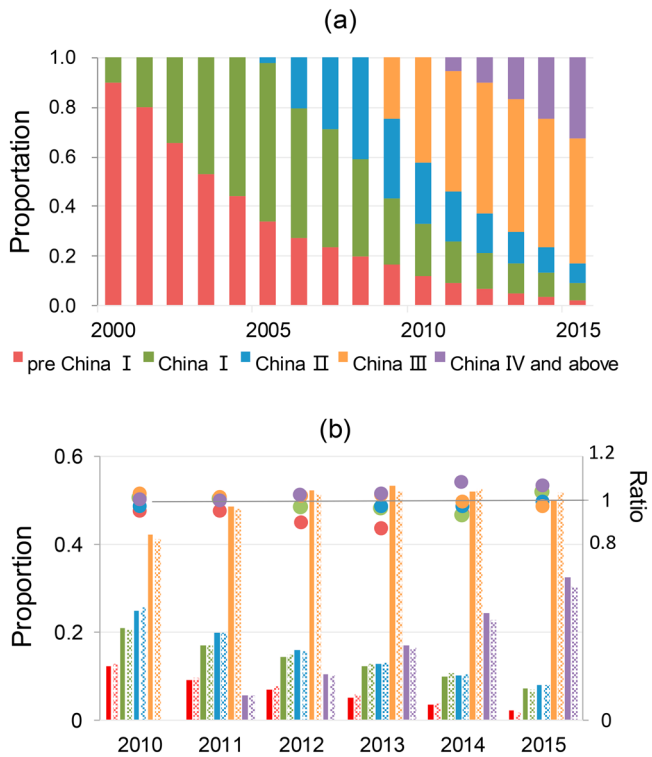

Figure 2. (a) Proportions of the vehicular population with different emission standards and (b) the comparison between our estimated proportion of emissions limits and the data from China MEP. Solid and shaded columns represent the proportion of the vehicular population at different limits (red: pre-China I, green: China I, blue: China II, orange: China III, and purple: China IV and above) in this study and in the MEP data, respectively. Symbols represent the ratio of this study/MEP data, and the distribution ranges from 0.91 to 1.1.

biomass burning, and nonroad mobile sources. The emissions of every pollutant were calculated with the following equations:

$$E_v = \sum \left(EF_{t,l,s} \times VMT_t \times P_{t,s}\right) \tag{1}$$

$$E_i = \sum_j \left\{ \left(A_{i,j} \times EF_{i,j}\right) \times \sum \left[\left(1 - f_{i,j}\right) + f_{i,j} \times \left(1 - \eta_{i,j}\right) \right] \right\} \tag{2}$$

where E_v is the vehicular emissions (Mt) of the four pollutants, t is the vehicle category, l is the driving cycle, s is the vehicular emissions standards at some stage, $EF_{t,l,s}$ is the EF (g/km) of vehicles in category t under driving cycle l at emissions standard s, VMT_t is the annual vehicle mileage traveled (km) for category t, $P_{t,s}$ is the population of vehicles for category t at emissions standard s, i is the source category, j is the sector under source i, E_i is the emissions (Mt) from other sources besides vehicles, $A_{i,j}$ is the activity of sector j, $EF_{i,j}$ is the EF of sector j, $f_{i,j}$ is the promotion rate of certain control technology, and $\eta_{i,j}$ is the removal efficiency of certain control technology.

2.2.2. Vehicular Emissions

Vehicular emissions for SO_2, NO_x, CO, and NMVOCs were calculated based on the modeled EF, annual average mileage, and the population of different vehicular fleets, using the COPERT 4 model (version 11.3; http://www.emisia.com/). This model was developed by the European Environment Agency and updated in June 2015 to estimate atmospheric pollutant emissions from on-road transportation. Compared with MOtor Vehicle Emission Simulator (MOVES2014a) models, COPERT is more suitable for estimating China's vehicular emissions because the dominant vehicle manufacturing technologies in China originated from European technologies, and vehicle emissions regulations implemented in China are almost the same as those performed in Europe (Cai & Xie, 2007). In addition, the COPERT model is a better fit for a country with different emissions regulation and deficient transportation data.

The calculation principle is shown in equation (1). There are some differences between European and Chinese vehicular classification standards; thus, we reclassified the vehicle population based on fuel type, function, and loading and obtained the fleet configuration under the European system. We chose 20 out of 266 available vehicular subsectors (see Table S2 in the supporting information). The population data were obtained from China Automotive Industrial Yearbook (China Automotive Technology Research Center, 1986, 1988, 1991, 1993–2015), China Statistical Yearbook (Editorial Department of China Traffic Yearbook, 1986–2016), Fifty-year Compilation of China Traffic Statistical Data (National Statistical Bureau, 2000).

Limits and Measurement Methods for Exhaust pollutants from vehicles began to release from 1993 by the Ministry of Environment Protection (MEP) of the People's Republic of China, which are generally consistent with the corresponding European standards. At present, there are multiple types of vehicles with different emissions regulations, rang from China I to China VI Emission Standard, on the road in China; in addition, vehicle that does not meet China I Emission Standard is called pre-China I in this study. China V Emission Standard was released in 2013 and implemented in 2017 nationwide; therefore, the vehicle population of China V and China VI emission standard had a quite low proportion before 2015, and it is included in "China IV and above." The implementation timeline of emissions standards, which mostly come from the Compilation Guide for the Air Pollutant Emission Inventory (hereafter called CGEI; MEP, 2014b) of on-road transportation (see Table S3 in the supporting information for the detailed timeline); otherwise, emissions standards for gasoline passenger cars from 1994 to 1999 refer to GB484-1993 (released in 1993 by MEP). We redistributed the vehicular population based on the implementation time of limits, vehicle renewal rate, and scrappage rate and compared our results with statistical data from MEP (Figure 2) to reduce the uncertainty of vehicular estimation. The proportion of the vehicular population with different limits in this study was largely consistent with the MEP data.

Other important parameters include average speed, fuel consumption, sulfur content, annual temperature, relative humidity, Reid vapor pressure, and VMT. Reid vapor pressure and average speed were estimated based on the literature (Bo et al., 2008; Cai & Xie, 2007). The VMT data were collected from the CGEI for on-road transportation. Annual temperature and relative humidity data were from China Meteorological Yearbook (China Meterological Administration, 1986–2016). Sulfur content and fuel consumption were from the annual Limits and Measurement Methods for Exhaust pollutants from vehicles, CESYs, and the China Automotive Industrial Yearbook (China Automotive Technology Research Center, 1986, 1988, 1991, 1993–2015).

2.2.3. Determination of EFs

Emissions from other sources besides vehicles were calculated based on time-varying EFs and activities among different sectors, fuel types, products, and emissions control technology (equation (2)). The EFs were determined as indicated in Figure 1. Our priority was to use locally measured EFs, which were collected from related literature (Ge et al., 2001; Lei et al., 2011; Li, 2007; Li et al., 2007; Liu & Zhu, 1997; Sun et al., 2004; Tian et al., 2011; Tsu, 2001; Wang & Zhang, 2008; Wang, Wei, et al., 2009; Yuan et al., 2005; C. Zhang, 2008; Q. Zhang et al., 2007; Zhao et al., 2012). Because those EFs are very limited, the EFs from CGEI were also adopted as estimations. In addition, we introduced some EFs from other countries from the literature and from databases (Jenkins et al., 1996; Kadam et al., 2000; Streets et al., 2006) and corrected these values based on a series of national limits of pollutant emissions (Ministry of Petroleum Industry of China, 1959; National Petroleum and Chemical Industry Bureau, 1992, 2000; National Technical Committee for Standardization of Paint and Pigment, 2001, 2008, 2009) and technical levels (see Table S1 in the supporting information for the full list of EFs). In general, the technical levels in China were approximately 10 years behind those in developed countries. Therefore, some EFs in a given year in China were selected from the EF data 10 years previous for the same sources in the foreign databases, such as AP-42 (USEPA, 1995), rather than selecting the updated EF data representing the current emissions levels in the developed countries (Bo et al., 2008). For some important sources, EF corrections are as follows.

Changes and corrections to EFs for important emissions sources were driven by government policies, technical improvements, and control measures, and the details are discussed by source in the following sections. Onroad EFs changed with the implementation of national limits (see Table S3 in the supporting information), whereas EFs of biomass burning were assumed to remain constant over the years.

2.2.3.1. Fossil Fuel Combustion and Industrial Processes

Technical improvements and exhaust control measures play an important role in reducing EFs. Taking NO_x as an example, the EF of uncontrolled NO_x from a coal-fired power plant was assumed to be 9.95 g/kg coal (Kato & Akimoto, 1992). Low-NO_x combustion technology, mainly low-NO_x boilers (LNBs), were used to reduce NO_x production in China starting in the 1980s, and the EF was estimated to be 5.97 g/kg coal according to the removal rate of 40% (C. Zhang et al., 2008). The penetration of LNB technology increased from 29% in 2000 to 65% in 2010. After 2005, fuel gas denitrification devices, including selective catalytic reduction (SCR, accounting for 94%) and selective noncatalytic reduction (SNCR), were used to reduce exhaust gas from boilers over the years according to China Association of Environmental Protection Industry (CAEPI, 2015). The NO_x EF of SCR boilers was estimated to be 1.393 g/kg coal based on a removal rate of 86% (Zhao, Wang, et al., 2013). The penetration of SCR technology increased from 1% in 2004 to 70% in 2015, according to C. Zhang et al. (2008) and China Electric Power Yearbook (China Electricity Council, 1993–2016).

Regarding industrial boilers, the government began to control NO_x emissions in the 1990s through LNB technology. The NO_x EF of coal-fired industrial grate boilers was 5.6 g/kg coal (Zhao, Wang, et al., 2013), and the LNB factor was 3.36 g/kg coal based on a removal rate of 40%. In addition, high-efficiency and low-pollution industrial circulating fluidized bed boilers emit 1.2-g NO_x per kilogram coal. The penetration of fluidized bed, automatic fire grate stoker, and manual grate stoker in industrial boilers are 0%, 50%, and 50% before 1980; 5%, 58%, and 37% in 1990; 10%, 80%, and 10% in 2000; and 23%, 75%, and 2% after 2010, respectively, according to the statistics from China Electrical Equipment Industry Association and China Machinery Industry Yearbook.

As for the cement industry, shaft kilns, cement rotary kilns and new dry process are the mainstream technology of cement production at present, and the NO_x EFs of cement production is based on local test results (C. Zhang et al., 2008). The NO_x EF of new dry process cement with SNCR was calculated at 50% removal rate. Changes in other EFs, such as power plant boilers, coking, and iron and steel production for CO, are also included in Table S1 in the supporting information.

2.2.3.2. Solvent Utilization

The VOCs EFs of asphalt paving were estimated as 150, 58, 41.8, and 35.5 g/kg asphalt in 1949–1991, 1992–1999, 2000–2010, and 2011–2015, respectively, according to Petroleum and Chemical Industry Standards SYB 1811-59, SH 0522-92, SH 0522-2000, and the CGEI. Similarly, we determined the EFs of interior and exterior wall coating from the Standards for Harmful Content Limits in Coating GB18582-2001, GB18582-2008, GB24408-2009, and the CGEI.

2.2.3.3. Nonroad Mobile Sources

There were five subsectors under this emissions source, and the EFs of air transport and railway diesel locomotives were assumed to be unchanged based on data from the CGEI for nonroad mobile sources. Regarding construction and agricultural machinery, the EFs were divided into four stages according to different emissions levels. For example, the VOCs EFs for construction machinery were 8.14, 7.02, and 5.77 g/kg fuel in 1949–1990, 1991–2000, and 2001–2008, respectively, based on European Monitoring and Evaluation Programme (2013), and 3.39 g/kg fuel in 2009–2015, based on the CGEI for nonroad mobile sources. The NO_x EFs of ship transportation were 47.6 g/kg fuel in 1949–2000 based on the CGEI and 25 and 23 g/kg fuel in 2001–2005 and 2006–2015, respectively, according to C. Zhang et al. (2008).

2.2.4. Activities

Most activities were directly collected from governmental statistical yearbooks. Additionally, some were estimated from related parameters, such as biomass burning and statistical data missing from the early years, as follows.

2.2.4.1. Industrial Processes

This source had four levels of classification, involving many sectors and subsectors. The activities of this source, such as energy consumption in the industrial process, material consumption and production, were mostly collected from China Statistical Yearbook (National Bureau of Statistics, 1981–2016), China Market Yearbook (China Strategic Research Ltd., 1999–2016), China Industry Statistical Yearbook (National Bureau of Statistics, 1990–2016), China Industrial Economy Statistical Yearbook (National Bureau of Statistics, 1994–1984, 1986, 1988–1995, 1998, 2001–2016), China Chemical Industry Yearbook (China Petroleum and Chemical Industry Association, 1994–2016), World Chemical Industry Yearbook (China Petroleum and Chemical Industry Association, 1984–1993), and the China Compendium of Statistics (1949–2008; National Statistical Bureau, 2010). For lack of statistical data in some sectors and from some years, the missing data were estimated in the industrial process (see Text S2 in the supporting information) and also applicable to other sources.

2.2.4.2. Biomass Burning

This source was classified into biofuels and open burning of agricultural residue. The activity of biofuels, including the consumption of biogas, cornstalks, and firewood as household fuel, were obtained from the CESYs, China Rural Statistical Yearbooks (National Bureau of Statistics, 1985–2016), and China Agriculture Yearbook (Editorial Department of China Agriculture Yearbook, 1980–2016). For lack of direct statistical data on the amount of open biomass burning, we used the estimation method from Bo et al. (2008; see Text S2 in the supporting information).

2.2.4.3. Fossil Fuel Combustion

Fossil fuel consumption of electric and industrial sectors was collected from the CESY, China Statistical Yearbook, and China Compendium of Statistics (1949–2008). The energy consumption of industrial boiler was calculated to be total energy consumption of industrial sector minus that of industrial process. Shan et al. (2016) found that there was a 20% gap between the aggregated energy consumption from 30 provinces and national consumption, when they calculated the provincial CO_2 emissions in China. Then NBSC corrected the energy data in 2015. Total energy consumption increased from 54 million tons of coal equivalent (Mtce) in 1953 to 4,299 Mtce in 2015, and the proportion of coal consumption decreased from 94.3% to 63%. In 2015, the coal consumption from thermal power, heating, industrial boiler, and resident was 17.93, 2.4, 9.08, and 0.93 Mt, respectively, which is basically consistent with statistical data from Shan et al. (2018). The missing data in the early years were estimated by per capita gross domestic product (GDP).

2.2.4.4. Solvent Utilization

There was little direct statistical data on the activity of this source, and thus, we estimated it from related parameters. The production and consumption of paint and printing ink were basically the same. The consumption of asphalt for paving was equal to the production of petroleum asphalt multiplied by the paving proportion. The activity of office supplies equaled the number of cultural and sports goods enterprises. The dry-cleaning activity was the consumption of trichloroethylene and vinyl chloride. The statistical data were obtained from

the China Statistical Yearbook, China Market Yearbooks, China Light Industry Yearbook, China Building Materials Industry Yearbook (Editorial department of China Building Materials Industry Yearbook, 1981–2016), and China Industrial Economy Statistical Yearbook.

2.2.4.5. Nonroad Mobile Sources

This source was classified into construction machinery, agricultural machinery, ship transport, air transport, and railway diesel locomotives, and the corresponding activities were diesel consumption in the construction industry, diesel consumption of agricultural machinery, ship fuel oil consumption, aircraft landing and takeoff cycle, and railway diesel engine fuel consumption, respectively. Activities came from the China Building Machinery Industry Yearbook, China Agricultural Machinery Industry Yearbook (Editorial Department of China Agricultural Machinery Industry Yearbook, 1987, 1992–2016), Statistical Bulletin of Highway and Waterway Transportation Industry (Ministry of Transport of the People's Republic of China, 2000–2016), and the Statistics Bulletin of Civil Aviation Airport Production (Civil Aviation Administration of China, 1994, 2002–2016) or were estimated; the estimation method referred to the CGEI for nonroad mobile sources.

2.2.5. Uncertainty Analysis

In this work, annual emissions uncertainties were estimated by a Monte Carlo approach with Crystal Ball software (USEPA, 2007). The uncertainty of activity was assessed by assuming a fixed coefficient of variation (CV; 10% for official statistics with normal distributions; 50% for estimated values with log-normal distribution). Log-normal distributions were applied for VOCs EFs with CVs of 50% for domestic databases and 100% for foreign databases. Triangular, uniform, normal, and log-normal distributions were used for SO_2, NO_x, CO EFs with CVs referenced in the literature (Lu et al., 2011; Wang et al., 2014; Yu et al., 2012; Zhao et al., 2011, 2012). All input parameters, including probability distributions, means, and CVs were incorporated into a Monte Carlo model, and 100,000 valid simulations were run to characterize the uncertainty, obtaining the probability density function, mean, median, 2.5th percentile, and 97.5th percentile values of output emissions. The estimated uncertainty was expressed as 95% confidence intervals and is shown as the shadow in Figure 6.

2.2.6. Provincial Allocation

We estimated the provincial emissions of four pollutants in 2015. Most province-level statistical data were gathered from statistical yearbooks of each province and regional statistical table of national yearbooks mentioned above. Parts of activity data in industrial process and solvent usage were distributed national statistics to provinces by industrial GDP, energy consumption, and other relevant parameters. In addition, production-to-residue ratio of crop in each province was determined by the report of the Ministry of Agriculture. The provincial desulfurization rate and denitrification rate in power plant were gathered from the data of China Electric Power Association. Vehicular emission control varied with the implementation time of emission standards.

2.3. Speciated VOCs and OFP Calculation

Most source profiles used in this study (68 sources) are from a VOCs profiles database compiled by Mo et al. (2016) and included sampling conducted in China in recent years. The other 34 source profiles, of which VOCs emissions accounted for 25.1% and local measurements were lacking, were retrieved from the SPECIATE database v4.5. Speciated VOCs were calculated by multiplying emissions by their corresponding weight percentage:

$$E_{i,S} = E_i \times p_{i,S} \tag{3}$$

where $E_{i,S}$ is the VOCs emissions from source i for chemical species s, $p_{i,S}$ is the weighted percentage of chemical species s from source i, and i is the chemical species (e.g., ethene or benzene). There are approximately 103 individual species in this integrated profile.

The OFPs of different species can be calculated by maximum incremental reactivity (MIR) and have been widely used to assess the key species and sources of ozone formation and to guide cost-effective ozone control measures (Li et al., 2014; Wu & Xie, 2017; Zheng et al., 2009). Individual species are calculated by multiplying emissions by the corresponding MIR value:

$$OFP_{i,S} = E_{i,S} \times MIR_S \tag{4}$$

where $OFP_{i,S}$ is the OFP of chemical species S from source i; and MIR_S is the MIR for chemical species S. The updated MIR values were from Carter (2008).

2.4. Scenario Analysis

The future VOCs emissions in 2018 or 2020 were predicted based on the 2013 inventory under three scenarios: (1) a basic scenario based on the current energy infrastructure and economic development level (until the end of 2013); new VOCs control policies would not be released or implemented; current legislation and implementation status will be followed until 2020. (2) Emissions-based control policy scenario, in which major industrial VOCs emissions would be cut by 3.3 Mt from 2016 to 2018, following the control measures of the MIIT Plan. The other sources remain as in the basic scenario. (3) Reactivity-based control scenario aimed at high-OFP sources, such as gasoline vehicle exhaust, industrial boilers, biofuel burning, and chemical production. This would intensify the control measures, including low-VOC replacement materials, energy-saving strategies, and end-of-pipe treatment for major pollution sources based on the MIIT Plan. The other sources remain as in the basic scenario. Detailed information on these three scenarios is summarized in Table S4 in the supporting information.

2.4.1. Assumptions About Future Activity

A projection of energy consumption, GDP, and population was adopted from the International Energy Outlook 2017 (Energy Information Administration, 2017) in the reference case. The activity of fossil fuel combustion was derived from energy consumption by end use sector and fuel. Transportation-delivered energy was used to estimated vehicular population. The industrial production, solvent usage, and other activities were fitted by time series curve or GDP and population, and some solvent consumptions were collected from Wei et al. (2011). In the reactivity-based scenario, energy-saving strategies were taken into consideration, according to Energy Development Projects (2014–2020, [2014] No31 State Council) issued by the State Council. On the one hand, the proportion of coal consumption would be decreased to 62% to optimize the energy structure. On the other hand, natural gas, nuclear power, and other clean energy sources were assumed to be further developed and promoted; for instance, the proportion of natural gas would be increased to 10% in 2020. As for the industrial process, coke production would be decreased by 20% in 2020, and machine coking would replace indigenous coking completely, due to adjustments to the national industrial structure (Yang, 2016). Moreover, we assumed that water-based paint and environmentally friendly inks would gradually replace solvent-based paint and traditional inks in the emissions-based and reactivity-based scenarios, as shown in Table S4 in the supporting information.

2.4.2. Scenario Design of Control Measures

The Chinese government has realized that industrial processes and paint usage are the major sources of VOCs emissions, contributing more than 50% of VOCs emissions. The MIIT and Ministry of Finance cooperatively set specific control measures and aims, including more than 20% reductions in benzene, toluene, xylene, dimethyl formamide, and other solvents used in industrial processes and increasing the proportions of low/non-VOCs replacement materials to 70% for pesticides, 60% for paint, 70% for inks, 85% for adhesives, and 40% for tires. Under this scenario, VOCs emissions would decrease by 15.8%, and the reduction targets of the 13th (2016–2020) FYP would be successfully achieved, which was used as emission-based control policy scenario.

In the reactivity-based control scenario, we selected high-OFP sources and intensified the corresponding control measures on the basis of MIIT Plan while loosen the restrictions on other sources. Firstly, vehicular emissions, particularly gasoline vehicle exhaust, contributed 18.5% of the total OFP. Although the MEP has issued the most stringent limits on vehicular emissions (China VI) in the world, pre-China I, China I, and China II vehicles still account for 45% of the vehicle population, and discharge 74% of VOCs. Based on vehicle age (shown in Figure S1 in the supporting information), most pre-China II vehicles will reach retirement age in 2020; thus, we considered the elimination of old cars as a vehicular reduction method that is not included in the MIIT scenario. Second, biofuel burning contributed 19% of the total OFP; thus, we assumed that biofuel consumption would decrease by 20% in 2020 compared with the basic scenario. In addition, coal consumption in industrial boilers would decrease by 10%. Finally, we increased the penetration of control measures for other high-OFP sources, such as tire, asphalt and butadiene production, and architectural paint usage. The details of two policy scenarios were shown in Table S4.

3. Results

3.1. Emission Trends Overview

The SO_2, NO_x, CO, and VOCs emissions trends and source contribution were shown in Figures 3 and 4. Annual emissions were listed in Table S5–S8, and the emissions at the provincial level in 2015 were shown in Table S9.

10.1029/2018EF000822

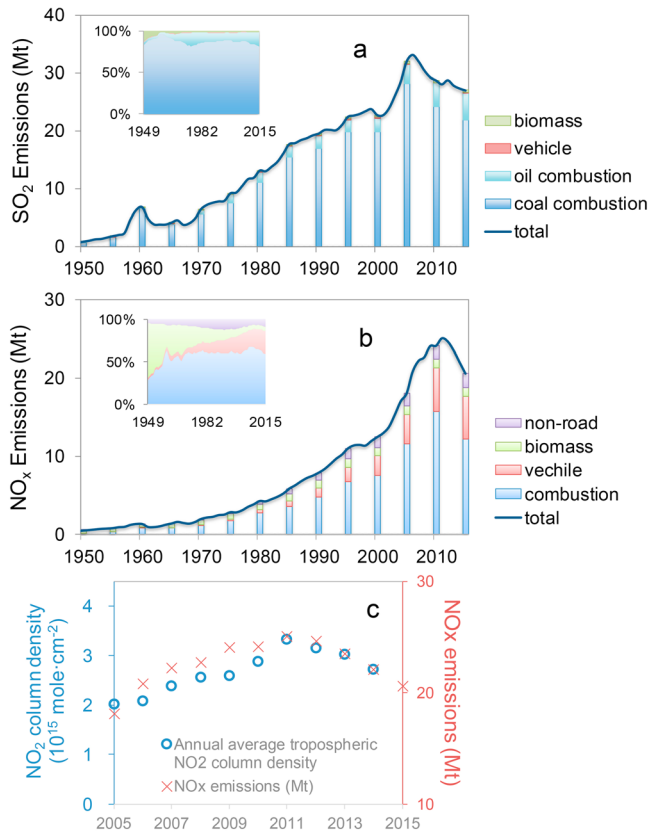

Figure 3. Anthropogenic SO_2 (a) and NO_x (b) emissions trends (Mt/year) and source proportion in China, 1950–2015. (c) Changes of tropospheric NO_2 column density in China and NO_x emissions during 2005–2015. VOC = volatile organic compound.

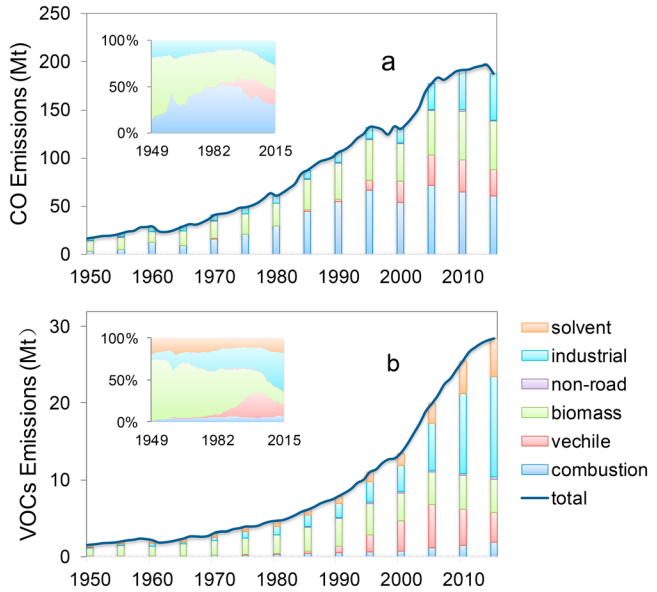

Figure 4. Anthropogenic CO (a) and VOCs (b) emissions trends (Mt/year) and source proportion in China, 1950–2015. VOC = volatile organic compound.

3.1.1. SO_2

Total SO_2 emissions in China were estimated at 27.06 Mt in 2015, and the contributions of fossil fuel combustion, on-road transportation, and biomass burning were 26.583, 0.08, and 0.40 Mt, respectively (see Table S5 in the supporting information for annual SO_2 emission data). Fossil fuel combustion, including power generation, heating, industrial boilers, and residential consumption, was the major source of SO_2 emissions, accounting for 98.4% (15.5% from oil combustion and 82.8% from coal combustion). Figure 3a show the historical trend of SO_2 emissions and the percentage contribution from each source. SO_2 emissions in China totaled 0.85 Mt in 1950, 13.14 Mt in 1980, 22.79 Mt in 2000, and 27.06 Mt in 2015. Total emissions increased significantly, but the contributions of the three sources were basically unchanged and fuel combustion accounted for more than 90% throughout the considered period. They historical trends can be divided into five periods.

From 1949 to 1999, SO_2 emissions increased by a factor of nearly 40, from 0.66 Mt in 1949 to 23.81 Mt in 1999, which was driven by the rapid growth of energy consumption. Emissions peaked from 1958 to 1960, corresponding to the Great Leap Forward in China. The steel and iron movement resulted in increased coal consumption and an increase in the emissions percentage of fossil fuel combustion from 1958 to 1960. During the three subsequent years of difficult economic times, SO_2 emissions rapidly decreased and remained at low levels for a long time. Starting in 1970, SO_2 emissions increased linearly with an annual growth rate (AGR) of 4.6%, consistent with the AGR of other bottom-up inventories (3.0–6.8%; Lefohn et al., 1999; Smith et al., 2011; Streets et al., 2000; Su et al., 2011; Wang et al., 1996). The percentage of vehicular emissions increased with the growth of the vehicle population until 1995, when the MEP began to implement nationwide vehicular emissions standards and the percentage of vehicular emissions gradually decreased.

From 1999 to 2001, SO_2 emissions decreased by 4.4%, from 23.81 to 22.78 Mt. Energy consumption decreased slightly, partly due to the impact of the Asian economic crisis but also because the government delimited acid rain and SO_2 control areas and took a series of measures to reduce SO_2 emissions.

After 2000, SO_2 emissions increased rapidly from 22.79 Mt in 2000 to 33.20 Mt in 2006, with an AGR of 6.47%. Our estimated emissions based on fuel combustion are higher than the official results from the MEP (1989–2014), but this growth rate is in good agreement with other inventories (5.5–8.7%; Lu et al., 2010, 2011; Smith et al., 2011; Su et al., 2011; Q. Zhang et al., 2009). SO_2 emissions peaked during 2005–2007, with the rapid economic boom and an average GDP growth rate of 12.7%. The percentage of vehicular emissions increased with the growth of the vehicle population until 2000. When the MEP began to implement nationwide vehicular emissions standards and limited sulfur content in gasoline and diesel, the percentage of vehicular emissions decreased rapidly. The sulfur content of gasoline in China V standard is capped at 10 mg/kg, therefore the SO_2 emission from vehicles is at a low level, less than 1% of that from coal combustion.

After 2006, SO_2 emissions began to decline significantly. Although energy use had been consistently growing since 2000, the government set strict emissions reduction targets during the period of the 11th (2006–2010)

FYP, which called for SO_2 emissions to be reduced by 10%. We estimated that SO_2 emissions decreased by 12.2%, from 33.2 Mt in 2006 to 28.79 Mt in 2010, with an FGD penetration rate of 12–83%, and the phase-out of small, high-emitting power-generation units. In addition, we found tropospheric SO_2 vertical column densities (VCDs) began to decline in 2007–2008 (Itahashi et al., 2012; Xia et al., 2016). During 2010–2015, SO_2 emissions leveled off as energy consumption continued to rise. The penetration of FGD installation in power plant tended to be saturated, but the raw coal washing rate remained at a low level.

3.1.2. NO_x

NO_x emissions in China were estimated at 20.6 Mt in 2015, and fossil fuel combustion, on-road vehicles, biomass burning, and nonroad mobile sources contributed 12.13, 5.52, 1.12, and 1.82 Mt, respectively (see Table S6 in the supporting information for annual NO_x emission data). The total Chinese NO_x emissions in this work (22.2 Mt in 2007 and 24.1 Mt in 2010) were lower than those reported by Li, Zhang, et al. (2017; 23.7 Mt in 2006 and 29.1 Mt in 2010), Zhao, Wang, et al. (2013; 26.1 Mt in 2010), and Zhao, Zhang, and Nielsen (2013; 23.8 Mt in 2007 and 28.8 Mt in 2010), but higher than the MEP results (17.98 Mt in 2007 and 24.043 Mt in 2011), as shown in Figure 6b.

Among the four sources, fossil fuel combustion was the primary source of NO_x emissions, and coal-fired boilers for power generation, industry, and residential sources contributed 43.8%, 28.6%, and 1.4% to this source, respectively, in 2015. In addition, 15.4% was contributed by coal consumption in the cement industry, and the remaining 12.9% was from oil, coal gas, liquefied petroleum gas (LPG), and natural gas combustion. Regarding on-road transportation, 80% of NO_x emissions came from diesel vehicles, and heavy-duty trucking was the dominant subsector, contributing 45.4% of vehicular emissions in 2013, consistent with the annual Report of Motor Vehicle Pollution Control in 2013 by the MEP (2014a). Although nonroad mobile sources were a small fraction of NO_x emissions, they emitted more NO_x than CO and VOCs, as diesel fuel and fuel oil were still the dominant energy sources used in nonroad engines.

Figure 3b show the historical trends and the contributions of the four sources, revealing that China's NO_x emissions increased from 0.52 Mt in 1950 to 4.30 Mt in 1980, at an AGR of 7.3%, and then increased by 5.46 times from 1980 to 2015. From 1949 to the present, NO_x emissions from biomass burning did not change much, but its proportion of total emissions decreased annually. By contrast, nonroad emissions increased, while the percentage remained at about 10%. The emissions and proportion of on-road vehicles kept rising annually. Fossil fuel combustion remained the dominant source of emissions and was also the focus of emissions reductions by the Chinese government.

Before the 1990s, although NO_x emissions were basically uncontrolled, total emissions only increased by 10.9 Mt from 1949 to 1998 because energy consumption increased slowly due to the low economic level. Emissions even declined in 1998 owing to the Asian economic crisis. However, total energy consumption increased sharply from 1,361 Mtce in 1998 to 3,870 Mtce in 2011, with an AGR of 8.4%, and NO_x emissions increased by 13.7 Mt, with an AGR of 6.2%. In the 1990s, a series of air pollution control policies and technological improvements were put into effect to reduce NO_x emissions, and these slowed down the rate of emissions growth under high energy consumption. Because fossil fuel combustion was the most important source for emissions mitigation, multiple measures were taken to control this source of emissions. Up until 2013, more than 90% of power plants and 60% of industrial boilers were equipped with LNBs, and the percentage of power plants with SCR was 50% in 2013 and reached 75% in 2015. The SNCR generation rate of cement kilns increased from 9% in 1999 to 76% in 2015. Tighter emissions standards for on-road transportation and off-road mobile sources were implemented step by step. The Chinese government set a target of a 10% reduction of NO_x emissions during the 12th (2011–2015) FYP period and has succeeded in reducing NO_x emissions by 18.6%, according to the China Environmental Status Bulletin by MEP (1989–2014).

Furthermore, the "inflection point" of emission has always attracted much attention, but few studies presented the inflection point of NO_x emission. In order to evaluate the NO_x emission trend in recent year, we compared the change ratio of emission and satellite observation data (Yu et al., 2017) during the period of 2004–2015 and found that the trend of annual average tropospheric NO_2 VCDs was in good agreement with our NO_x estimate (as shown in Figure 3c). Both of them all peaked in 2011. Annual average tropospheric NO_2 VCDs over mainland China increased from 2005 to 2010, with an AGR of 7%, compared with NO_x emissions' AGR of 5%. When multiple control measures were taken into effect during 12th FYP, NO_2 VCDs decreased with an AGR of 4.9% in 2011–2014, compared with NO_x emissions' AGR of 4% in 2011–2015. Tropospheric

NO_2 over mainland China from 2005 to 2014 using vertical column density data sets retrieved from the Ozone Monitoring Instrument (OMI) in the research of Yu et al. (2017).

3.1.3. CO

Total CO emissions in China were estimated at 188.8 Mt in 2015, and the contribution of coal combustion was the largest (31.8%), followed by biomass burning (27.3%), industrial processes (24.6%), on-road transportation (15.6%), and nonroad mobile sources (0.6%; see Table S7 in the supporting information for annual CO emission data). The EFs of liquid and gaseous fuels is quite low for complete combustion; therefore, we took coal and biomass as the main fuels for the combustion source. For the on-road transportation category, gasoline and diesel vehicles contributed 25.8 and 0.98 Mt, respectively; thus, incomplete combustion of coal, biomass, and gasoline is recognized as the major CO emissions source (72.5% of the total anthropogenic emissions) in China.

As shown in Figures 4a, China's CO emissions increased from 16.9 Mt in 1950 to 188 Mt in 2015, with an AGR of 3.8%. The main contributing source switched from biomass burning to coal combustion and industrial processes. Unlike SO_2 and NO_x, the government has not set a nationwide CO emissions reduction target, but China has successfully improved combustion efficiency and implemented emissions control regulations aimed at many sectors. Total CO emissions have been relatively stable since 2005, with an AGR of 1.24%, despite the fast growth of energy consumption and industrial production; this trend is consistent with the AGRs of 1.55% reported by Zhao, Zhang, and Nielsen (2013) and 1.82% reported by Yumimoto et al. (2014; shown in Figure 6c).

Among the five sources, biomass burning has always been an important CO emitter, and 70–90% of emissions from this source were released by biofuel-fired domestic boilers due to their low combustion efficiency. Before 1972, biomass burning was the largest source of CO in China, accounting for more than 40% of total emissions, and continuously increasing year after year. However, coal combustion replaced it as the largest CO emitter with the rapid growth of coal consumption after 1972, and industrial boiler emissions were much higher than those from power plants. Brick making and cement kilns contributed most of the CO emissions from coal-fired industrial boilers, collectively accounting for more than 53.7% of coal-fired emissions. Power plants emitted less than 3.0% of the total CO due to the high combustion efficiency of pulverized coal furnaces. The phase out of old brick and cement kilns and the increased ratio of bypass gas recycling effectively reduced CO EFs; thus, the proportion of coal-fired emissions decreased from 51.4% in 1990 to 32.4% in 2013.

With rapid industrial development, the share of industrial emissions increased from 11.1% in 2000 to 25.7% in 2015, and iron and steel production have been the largest emitter, accounting for more than 80% of industrial emissions, in agreement with the results of (Zhao, Zhang, & Nielsen, 2013). CO emissions from on-road transportation increased by 10 times from 1990 to 2014 with the rapid growth of the vehicle population, which increased 20 folds during this period, and passenger cars and motorcycles contributed more than 80% of vehicular CO emissions. This also shows that the growth rate of CO emissions slowed down, and vehicular CO in 2015 was 13% less than in 2009, mainly due to the application of strict emissions regulations on new vehicles and the forced elimination of old vehicles with high emissions levels. Additionally, nonroad mobile sources contributed less than 1% of total CO emissions.

3.1.4. Volatile organic compounds

Anthropogenic VOCs emissions in China were 28.43 Mt in 2015 and came from a wide variety of sources. Among the six major sources, industrial processes dominated the total emissions (13.08 Mt, 46%), followed by solvent use (17.6%), biomass burning (15.3%), on-road transportation (13.5%), fossil fuel combustion (6.5%), and nonroad mobile sources (0.98%; see Table S8 in the supporting information for annual VOCs emission data). In the subsectors of the industrial processes sector, the petrochemical industry was the main contributor, accounting for 59.1% of industrial VOCs, followed by coke production (18.8%). Pesticide use and asphalt paving were the dominant subsectors (34.9%) of solvent use, due to their high EFs and widespread use. Biofuel-fired domestic boilers and coal-fired industrial boilers were the largest contributors of emissions from biomass burning and fossil fuel combustion, contributing 81.85% and 79.36%, respectively, in 2013. As for on-road transportation sources, motorcycles and passenger cars made up 40.30% and 34.82% of vehicular emissions, respectively, due to their large population and high VMT. Among nonroad mobile sources, shipping transportation contributed 66.81% of the total emissions.

Historical emissions and the contributions from each source are shown in Figure 4b. Unlike the three pollutants described above, VOCs emissions generally increased from 1949 to 2015, and its AGR accelerated after China's economic reform and joining the World Trade Organization (WTO). From 1949 to 1977, the level of VOCs emissions in China was relatively low, with an AGR of 3.7%, and more than half of the population in China used agricultural waste and firewood to satisfy their energy needs. Therefore, biomass burning was the main emissions source in the early years, but its proportion has significantly declined in the last 20 years.

Since the economic reform, China's VOCs emissions increased rapidly from 4.31 Mt in 1978 to 13.49 Mt in 2000, with an AGR of 5.3%. The government had not instituted control measures before 2000; thus, the emissions from each source also exhibited increasing trends with steady rates. On-road emissions in particular showed strong growth with the increasing vehicle population, which increased by 40 times during this period and became the dominant contributor to VOCs emissions before 2005.

After 2000, although the government has not carried out a national emissions control and reduction plan for VOCs in China, some periodic emissions limits and standards have been undertaken for major individual sources, such as exhaust emissions from road vehicles, motorcycles, and nonroad engines, and the VOCs content of coatings, oil paint, and petroleum asphalt. Vehicular VOCs emissions declined from 5.64 Mt in 2004 to 3.85 Mt in 2015, with the rapid growth of the vehicle population, which increased by 2.4 times during this period, whereas the emissions from other sources were not well controlled and instead continued increasing. Total VOCs emissions doubled from 2000 to 2015, and the contribution from industrial processes increased from 25.5% in 2000 to 46.0% in 2015 due to the rapid growth of industries and less effective emissions control measures.

3.2. Speciated VOC Emissions and Reactivity

According to the 2013 VOCs emissions in China estimated in this study, 26.3% of total VOCs emissions were from aromatic compounds, whereas alkanes, oxygenated VOCs, and alkenes made up 21.0%, 20.3%, and 16.7% respectively. Figure 5a shows the top 10 mass-based species and their corresponding OFP contributions. Ethane contributed the largest fraction of emissions at 7% and the highest contributing aromatic species—toluene, styrene, and benzene—made up 6.6%, 5.2%, and 4.4%, respectively, of VOCs emissions. Moreover, ethane (3.9%) and acetaldehyde (1.9%) were the highest contributing alkane and oxygenated VOC species, respectively. However, the OFP-based contributions of VOC species were quite different from their emissions-based contributions. Alkenes and aromatics accounted for 48.1% and 30.5%, respectively, of total OFP. Ethane (18.3%), 1,3-butadiene (14.4%), propene (6.7%), toluene (6.6%), and m/p-xylene (6.0%) contributed more than half of the total OFP. The complete emissions and reactivity of 109 species are presented in Table S10 in the supporting information.

Sources of VOC emissions and OFPs and their contributions are shown in Figure 5b. Industrial production and biomass burning contributed the greatest proportion of emissions and OFP because ethane, formaldehyde, acetaldehyde, and propene accounted for a high proportion of biomass burning. In addition, the OFP contributions of paint, gasoline vehicle exhaust, fossil fuel combustion, and motorcycle exhaust exceeded the emissions contribution, and these sources were included in the priority control in the OFP-based scenario. Seventeen key species, shown in Figure 5c, contributed more than 80% of the total OFP, and eight of these were aromatic species. Ethene, a combustion product, was mainly produced from gasoline vehicle exhaust, industrial boilers, and biomass burning. 1,3-Butadiene was emitted by industrial processes, such as tire production (57.4%) and basic chemical production (39.0%). BTEX (toluene, xylene, ethyl benzene, and trimethyl benzene) were mainly generated from architectural paint and surface coatings. The sources mentioned above were also taken into consideration in the OFP-based scenario.

3.3. Regional Comparison

Asian emissions of the four considered air pollutants increased rapidly from 1970 to 2010 in many developing countries in Asia, and China's proportion increased and exceeded 30% in 2008 as shown in Figure S2. The proportion of SO_2 emissions in China was just 18% of the Asian total in 1970, and it rapidly rose to the highest level of 62% in 2005. After that, China's share decreased to 47% in 2010, owing to the promotion of SO_2 control measures in China. In recent years, China has become the number one emitter of CO in Asia, with a large amount of fuel consumption and a deficiency of control measures. The Chinese proportion of VOCs emissions was lower than that of the other three pollutants but grew rapidly from 6% in 1970 to 32% in 2010.

10.1029/2018EF000822

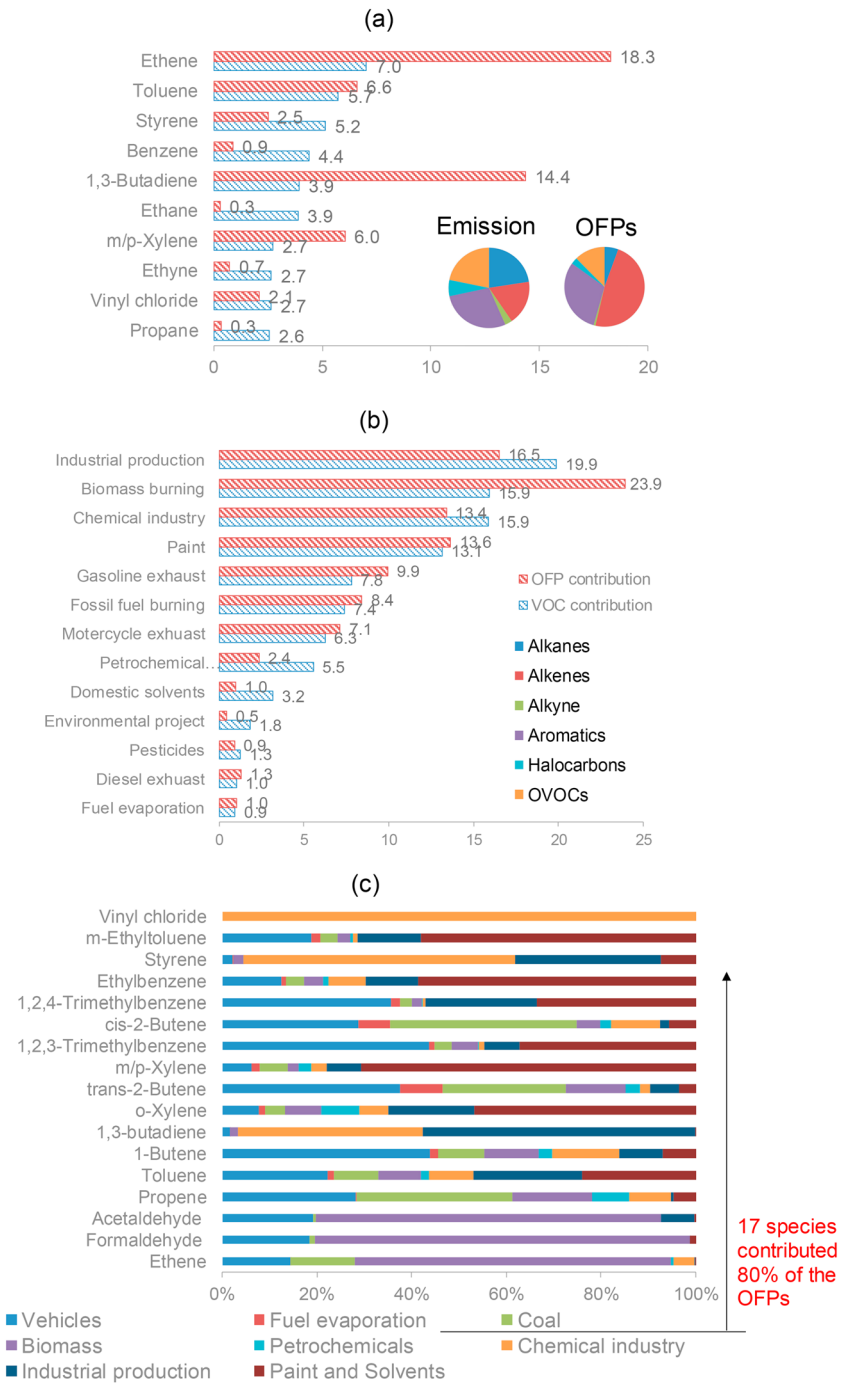

Figure 5. Key VOCs emissions-contributing and OFP-contributing species (a) and their sources (b). Seventeen VOCs species (c) contributed more than 80% of the total OFP. MEP = Ministry of Environment Protection of the People's Republic of China; OFP = ozone formation potential; VOC = volatile organic compound; OVOC = oxygenated VOC.

The global emissions growth rate was relatively flat because emissions have been falling gradually in recent decades in most developed countries, including the United States and European countries. However, the Chinese proportion of emissions exhibited large growth, from 2–8% in 1970 to 15–31% in 2010, and the sum of emissions from China and the United States accounted for about 20–30% of the total global emissions throughout the whole period. Emissions from the United States were several times that of China in 1970, but U.S. emissions have decreased steadily since the 1970s. Emissions from China increased to the same level as

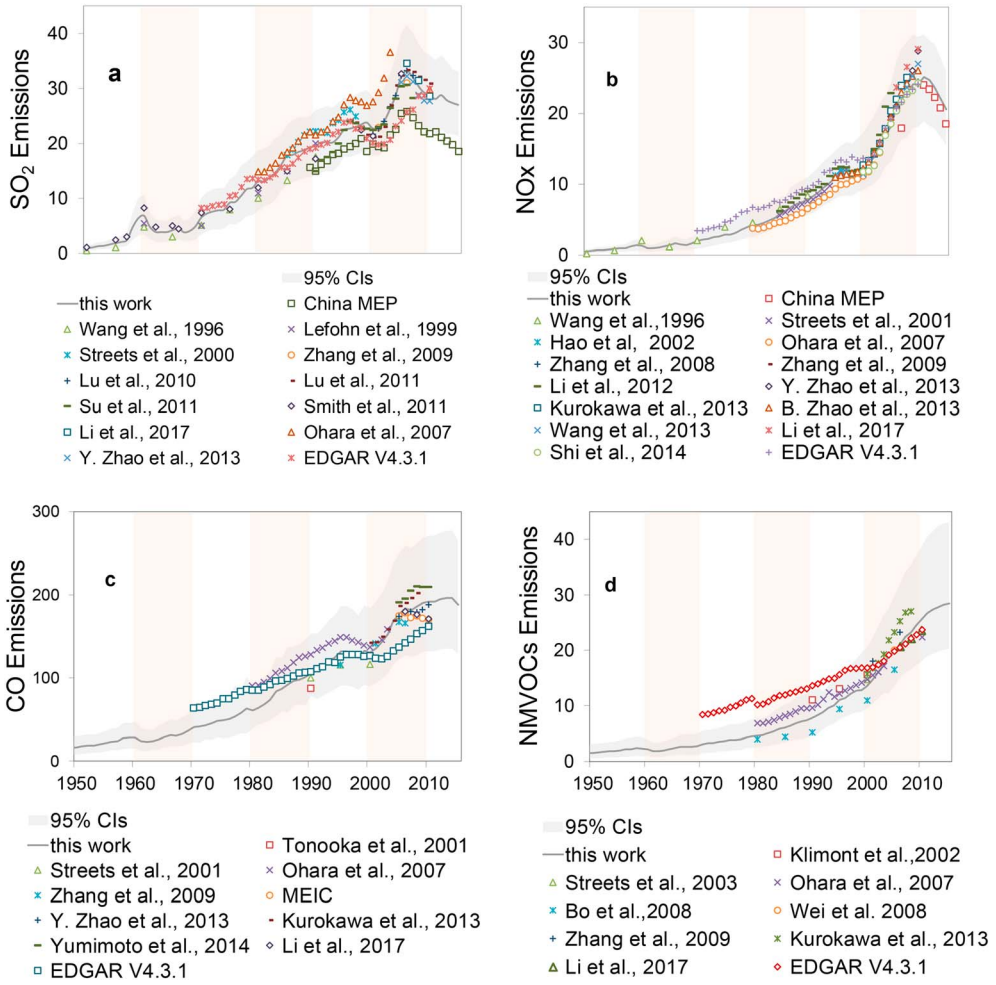

Figure 6. Comparison among multiple estimates of Chinese air pollutants emissions: (a) SO_2; (b) NO_x; (c) CO; (d) VOCs. The solid lines indicate our result, and gray areas indicate the uncertainty of our results (95% CIs). Other points indicate the estimated emissions from the China MEP or other studies. CI = confidence interval; MEP = Ministry of Environment Protection; NMVOC = nonmethane volatile organic compound.

that of the United States around 2000, and there was a shift in SO_2 and NO_x emissions with the pollution reduction policies and control measures implemented in China. VOCs emissions, however, still increased at an AGR of 2.79% after 2010 in China, and the reduction of total VOCs has become a major focus of governmental work at this stage.

4. Discussion

4.1. Comparisons With Other Estimates

To verify the historical emission trends, we compared our results with official statistics reported by the MEP (1989–2015 for SO_2, 2011–2015 for NO_x) and other researches, including regional and global inventories, such as Regional Emission inventory in ASia (REAS) (Ohara et al., 2007), Intercontinental Chemical Transport Experiment-Phase B (Q. Zhang et al., 2009), Multi-resolution Emission Inventory for China (http://www.meicmodel.org/), Emission Database for Global Atmospheric Research (EDGAR) v4.3.1 (Crippa et al., 2016), MIX inventory (Li, Zhang, et al., 2017), and Chinese emissions estimates, such as Bo et al. (2008); Su et al. (2011); Smith et al. (2011); and Zhao, Zhang, and Nielsen (2013). These estimated trends or single-year emissions levels are demonstrated in Figure 6. Both the government and the public pay more attention to vehicular emissions in China at present, and there are some significant differences among these estimates. Therefore, we compared vehicular emissions of NO_x, CO, and VOCs with previous estimates in Figure 7 individually and assessed the efficiency of national limits at different stages.

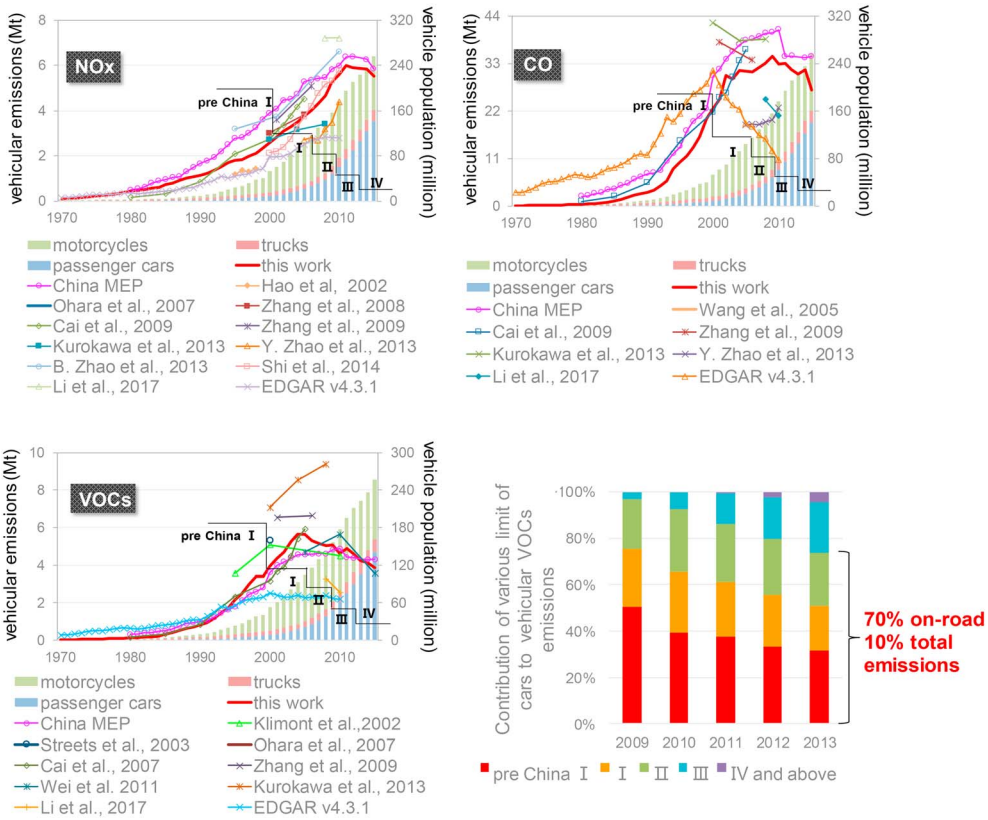

Figure 7. Comparison of different estimates of vehicular emissions: The columns represent the annual vehicular population; black folding lines represent different national limits (The implementation period of national limits differs by type of car; thus, we chose gasoline passenger cars as an example of typical limits.). The last picture presents the contribution of cars with different emission standard to vehicular VOCs emissions. Vehicles with China II, China I, and pre China I emission standard emitted 2.96 Mt of VOCs (74% of vehicular emissions and 10% of total emissions). MEP = Ministry of Environment Protection; VOC = volatile organic compound.

4.1.1. Total Emissions and Trend

As shown in Figure 6a, the total SO_2 emissions were consistent with most of the other inventories (Lefohn et al., 1999; Li, Zhang, et al., 2017; Smith et al., 2011; Q. Zhang et al., 2009; Zhao, Zhang, & Nielsen, 2013) but slightly lower than Ohara et al. (2007), Streets et al. (2000), and Lu et al. (2011). The reason for the inconsistency might be the exclusion of noncombustion industrial processes in this study, coupled with differences in some parameters used in each inventory. Sulfuric acid production, non-ferrous metal smelting and other noncombustion industrial processes were not included in our estimation, since those sources only contributed a tiny portion (2.1% in EDGAR v4.3.1) to total SO_2 emissions. The average FGD penetration was estimated as 9% in 2005, 78% in 2010 by Lu et al. (2011), 20% in 2005, and 86% in 2010 in this study; thus, the average coal EF reported by Lu et al. was higher than our estimates. In addition, it was reported that the REAS inventory overestimated the total SO_2 emissions by choosing high EFs (Lu et al., 2011; Smith et al., 2011; Q. Zhang et al., 2009). The underestimate of emission by Wang et al. (1996) might be the application of low sulfur content (1.12% in 1990) to 1950–1990. As the coal combustion was the dominant source of SO_2 emissions, the reason for the underestimation in EDGAR 4.3.1 might be the adoption of International Energy Agency (IEA) energy statistics. The coal supply value from National Bureau of Statistics (2,513 Mtce in 2005) were higher than that from IEA statistics (1,719 Mtce in 2005).

The agreement among various estimates of China's NO_x emissions is reasonably good before 2005 (Figure 6b; Crippa et al., 2016; Hao et al., 2002; Kurokawa et al., 2013; Li, Zhang, et al., 2017; Li & Li, 2012; National Research Council, 2010; Ohara et al., 2007; Shi et al., 2014; Streets et al., 2001; Wang et al., 2013; C. Zhang et al., 2008; Q. Zhang et al., 2009; Zhao, Wang, et al., 2013; Zhao, Zhang, & Nielsen, 2013). This is partly because these studies used the similar original EFs reported by Kato and Akimoto (1992), Streets and Waldhoff (1998), Streets

and Waldhoff (2000), Tian (2003), C. Zhang et al. (2008) and Compilation of Air Pollutant Emission Factors (AP-42) by United States Environmental Protection Agency (USEPA, 1995), and partly because most emissions sources were uncontrolled before 2005. From 2006 to 2010, many reduction measures, like LNB, SCR/SNCR, and boiler improvements, were applied to the major emitters: power plant boilers, industrial boilers, and cement kilns. The estimates from MIX and REAS2 were approximately 5–15% higher than our results, possibly due to underestimating the penetration of SCR/SNCR. Moreover, our estimate was quite close to those of Zhao, Wang, et al. (2013) and Shi et al. (2014), and the average control technology penetration of coal-fired power plants was estimated as 75% LNB and 12% SCR by Zhao, Wang, et al. (2013) and 73.9% LNB and 14% SCR in this study. Total NO_x emissions were estimated to decline both in this study and in statistics from the MEP during the 12th (2011–2015) FYP, but our estimates were higher than the official data by approximately 2 Mt. The reason for this inconsistency may be the inclusion of the contributions of nonroad, about 2 Mt, in our inventories, which were excluded in the official data.

As shown in Figure 6c, EDGAR v4.3.1 and REAS estimated higher CO emissions than our results owing to the overestimation of emissions from the residential sector before 2000; this sector contributed 65% in EDGAR and 60% in REAS from 1970 to 2000, compared with 35% in this study. As the residential coal consumption from the IEA are almost the same as those in the CESY (100 versus 94.9 Mtce in 1995), the reason for the inconsistency in this sector may be the differences in the EFs. Namely, the average coal-fired domestic EF, excluding biofuel combustion, of REAS (167 g/kg coal) is much higher than those tested in China by Zhang et al. (2000; 71.3 g/kg coal) and in this work (75 g/kg coal). Moreover, the rapid growth of CO emissions agreed well with most of the other inventories, with total energy consumption doubling from 2000 to 2006, except for EDGAR v4.3.1, and the total emissions in REAS2 were found to be overestimated after 2005 with high EFs (Li, Zhang, et al., 2017). A series of control measures, such as the phase out of old kilns and improved combustion efficiency, effectively reduced EFs for industrial sources during the 11th (2006–2010) FYP. Considering those implementations, the growth rate of CO emissions decreased significantly in the inventories of Zhao, Zhang, and Nielsen (2013), consistent with this work; total emissions even decreased by 4.8% in MIX because of the higher reduction in vehicular emissions.

As for VOCs, the rapid growth of industry and less effective emissions control measures led to an upward trend in all existing inventories (Bo et al., 2008; Klimont et al., 2002; Kurokawa et al., 2013; Li, Zhang, et al., 2017; National Research Council, 2010; Ohara et al., 2007; Streets et al., 2003; Wei et al., 2008; Zhang et al., 2009). The REAS inventory and Klimont et al. (2002) estimated higher VOCs emissions before 2000 than did this work because higher EFs and activities were used. Moreover, the VOCs emissions in the REAS inventory were estimated simply by extrapolating the results from 1995 and 2000, based on the estimation of Klimont et al. (2002) and Streets et al. (2003) and may have overestimated on-road vehicular emissions. Bo et al. (2008) presented approximately 10–30% lower VOCs emissions than this work because we added nonroad emissions and extended from 15 to 45 industrial sectors. The VOCs emissions in the REAS2 inventory are higher than those in most inventories, which seems to be due to its high EFs from Klimont et al. (2002) and Streets et al. (2003). On one hand, the 2008 vehicular emissions calculated in REAS2 was 9.48 Mt, which was much higher than our estimate of 5.65 Mt because of their higher adopted EFs based on the European experience in the mid-1980s. On the other hand, the EFs of combustion were higher than in other inventories. For example, the EF of biofuel-fired civil boilers was 12.93 g/kg fuel in REAS2, 5.3 g/kg fuel in our work, and 1.13–9.37 g/kg fuel was recommended by the CGEI. The estimates in the MIX inventory were slightly lower than our results due to lower vehicular and industrial emissions.

The uncertainties (expressed as the 95% confidence interval around the central value) are shown in Table 1. We can find the uncertainty among the four pollutants increases with source category, and NMVOCs emissions have the largest uncertainty due to the complexity of sources and the lack of EFs data, especially for industrial process and solvent usage. Uncertainties in fossil fuel combustion are less than other sources, which benefit from sufficient statistic data and available local EFs, and it is recognized in other studies (Q. Zhang et al., 2009; Zhao et al., 2011). The distribution of vehicular population under different standards in various vehicle fleet is the major uncertain for on-road emissions. Uncertainties in biomass burning emissions derived from the estimation of open burning activity.

4.1.2. Vehicular Emissions

Although there were not many differences among estimates of total emissions, ranging from −30% to 80% around our results, vehicular estimates ranged from several to several dozen times (Figure 7) due to various

Table 1
Uncertainties of Total Emissions by Sector in 2015

	Fossil fuel combustion	Vehicle	Biomass	Nonroad	Industrial	Solvent	Total
SO_2	26.58 (−38%, 42%)	0.08 (−39%, 44%)	0.40 (−52%, 83%)	—	—	—	27.06 (−22%, 25%)
NO_x	12.13 (−24%, 22%)	5.52 (−48%, 76%)	1.12 (−34%, 92%)	1.82 (−52%, 104%)	—	—	20.59 (−26%, 34%)
CO	60.82 (−44%, 44%)	26.92 (−57%, 109%)	50.66 (−54%, 110%)	1.24 (−63%, 148%)	48.36 (−33%, 36%)	—	188.00 (−31%, 41%)
NMVOCs	1.86 (−38%, 61%)	3.85 (−56%, 97%)	4.34 (−60%, 95%)	0.28 (−87%, 279%)	13.08 (−33%, 89%)	5.02 (−37%, 62%)	28.43 (−32%, 56%)

Note. The estimated missions are expressed as kilo metric tons (Mt). The percentages in the parentheses indicate the 95% confidence interval around the central estimate. NMVOCs = nonmethane volatile organic compounds.

emissions regulations and deficient transportation data. Our emissions trends were generally consistent with most of the inventories, but the most significant difference mainly existed in EDGAR and MIX. NO_x emissions from vehicles in 2010 were estimated at 5.58 Mt in this work, 7.22 Mt in the MIX, and 2.80 Mt in the EDGAR inventory. Estimation from EDGAR slumps after 2000 and becomes significantly lower than that of the other results, probably due to overestimating the effects of emissions regulation at an early stage. The results of the three calculations were compared with the official statistics reported by the MEP, and our calculations were consistent with those of the MEP (5.99 Mt).

The vehicle population in China increased by 10 times over the past 20 years and reached 250 million in 2015 (55% passenger cars, 8% trucks, and 37% motorcycles). It is estimated that new vehicles will exceed 100 million during the period from 2015 to 2020 according to the MEP; thus, vehicular emissions control is receiving greater public attention. Most of the estimated results indicated that vehicular emissions declined over the past 5 years; VOCs emissions from vehicles began to decline after the China II period, and CO and NO_x started to fall after the China IV period in this work, consistent with the MEP data. China IV vehicles have been promoted nationwide with low pollutants emissions, and 13% of the population in 2013 only emitted 4.4% of vehicular VOCs emissions. However, vehicles with China II, China I, and pre-China I emission standard accounted for 45% of the population and emitted 2.96 Mt of VOCs (74% of vehicular emissions and 10% of total emissions); therefore, decommissioning old vehicles is more important than strengthening emissions regulations.

4.2. Projection of NMVOCs Emissions

Figure 8 presents the estimates of future VOCs emissions under three scenarios and their corresponding OFPs. In the real situation, total VOCs emissions were 28.4 Mt in 2015, just between those under basic scenario (28.85 Mt) and OFP-based control scenario (28.1 Mt). In the basic scenario, the national NMVOCs emissions are projected to increase from 27.8 Mt in 2013 to 31.3 Mt in 2020 with AGRs of 1.74%. Industrial process and solvent utilization were still the major contributor to the growth of emissions due to the rapid development of industrial production and paint consumption. These projections show that the national VOCs emissions would decrease during 13th FYP under two policy scenarios and are projected to be 23.2 Mt in 2018 and 24.5 Mt in 2020 under the emissions control and reactivity control scenarios with AGRs of −4.3%, and −2.8%, respectively. Differences were observed in emissions-based and OFP-based source contributions, as shown in Figure 8a, and their proportions were 11% versus 7% from vehicles, 44% versus 50% from industrial processes, and 19% versus 14% from biomass burning. Industrial sources would contribute the largest emissions reduction, about 1.8 Mt compared with 2013 levels, followed by MIIT Plans under the emissions-based control scenario. Vehicular emissions would decrease by 2.56 Mt (compared with 2013 levels) under the OFP-based control scenario, and its proportion could reach as low as 7% in 2020 if the China VI standard (GB 18352.6-2016) is carried out nationally and pre-China II vehicles are completely eliminated.

Under the basic scenario, VOCs emissions and OFPs would reach the highest level in the historical trend if no more VOC control measures were taken after 2013. As for the two policy scenarios, the targets and effects differ, as shown in Figure 8b. The emissions-based control scenario focuses on 29 high-emission sources, such as basic chemical industry, synthetic fiber production, automobile paint, pesticide, reducing VOC emissions by 4.8 Mt between 2014 and 2018, with the benefit of a 13.3% reduction in OFP. In the reactivity-based

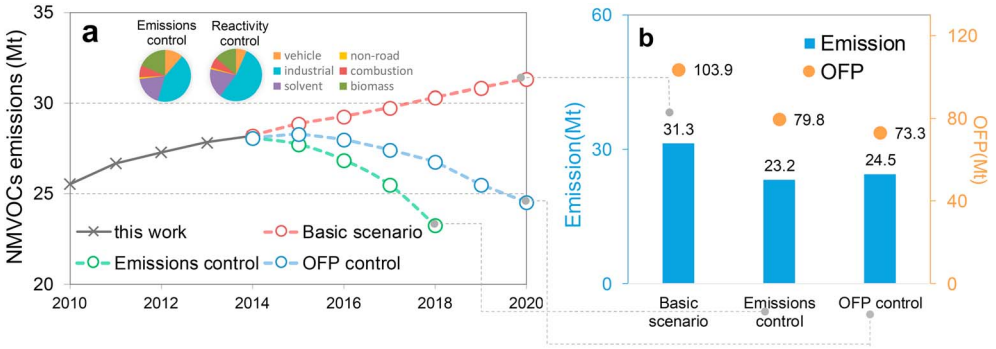

Figure 8. (a) Future VOCs emissions under three scenarios and (b) their corresponding OFPs. NMVOC = nonmethane volatile organic compound; OFP = ozone formation potential.

control scenario, 17 high OFP-contributing sources would be prioritized, including gasoline vehicle exhaust, tire production, architectural paint, and biofuel burning, which could lead to a 3.6-Mt reduction in VOCs emissions and a 20.4% reduction in OFP during the 13th FYP. Consequently, either of these two control measures could achieve the target of a 10% reduction in VOCs emissions during the 13th FYP. However, unlike other pollutants (SO_2 and NOx), VOCs pollution control cannot simply rely on total emission reduction. The purpose of scenario analysis in this study is to explore VOCs reactivity control strategy. Priority control of reactive VOCs species and sources will improve air quality more efficiently.

4.3. Environmental Policy Implication

Research on emission trends of major air pollutants can help policy makers develop strategies and policies for emission reductions and track the progress of those policies. While achieving the targets of emission reduction, environmental policies also favor energy structure adjustment and industrial production upgrading. At the initial period, end-of-pipe treatment was the primary means of emission control, such as the penetration of gas desulfurization and denitrification technology, and had remarkable effect on emission reductions. As the installation rate of FGD has been saturated in recent years, the driving force for continuous reduction gradually turned to energy structure adjustment and industrial upgrading. In heating, industrial, and agricultural processes, the government encouraged to use natural gas, nuclear power, and other renewable energy instead of coal and promoted the "coal to electricity, coal to gas" project in the nationwide to further optimize the energy structure. The national energy consumption of coal decreased by 107 Mtce from 2013 to 2016, while that of hydropower, nuclear power, and wind power increased by 36.4%. In addition, the task of emission reduction enhanced the determination of local governments to upgrade the industrial production. Some high-emission, low-GDP workshops, and small coal-fired boilers have been banned. In order to promote the VOCs emission reduction, enterprises are required to use low/non-VOCs replaced materials and implement clean production transformation in coating, ink production, petroleum refining, petrochemical, packaging printing, and other key industries.

According to the research on "Olympics blue" and "APEC blue," the reduction in emissions of secondary pollutant precursors (i.e., NO_2, SO_2, and VOCs) could have reduced the secondary aerosol and ozone formation in the atmosphere (Wang, Hao, et al., 2009; Zhang et al., 2018). In the 13th Five-Year Plan, the Chinese government included SO_2, NO_x, and VOCs into the total emission control and released a series of emission standards, reduction plan, and regulations. Multipollutant emission control could reduce the atmospheric pollution characterized by fine particles and ozone at present.

5. Conclusions

We identified the long-term trends of anthropogenic SO_2, NO_x, CO, and VOCs emissions in China from 1949 to 2015, with six major sources, based on a technology-based methodology, time-varying statistical data, and updated EFs to reflect the rapid increase in energy consumption and dramatic changes in technology penetration and control measures. The national SO_2 and NO_x emissions were confirmed to reach inflection point in 2006 and 2011, respectively. However, the installation rate of fuel gas desulfurization has been approaching

saturation in recent years. The promotion of alternative energy, LNB, and SCR technology would be the driving force for SO_2 and NO_x emission reduction for the next few years. In addition, we estimated speciated VOCs emissions and OFPs mainly using Chinese source profiles and found that gasoline vehicle exhaust, coal and biofuel combustion, architecture painting, and tire manufacturing, among others, were the major source categories contributing to ozone formation. A reactivity-based control strategy was proposed to achieve more OFP reduction (20%) than emissions-based control measures (13%), with less reduction in VOCs emissions (13%).

Acknowledgments
All emission data by source, EFs, and other information involved in this study are listed in the supporting information (Texts S1 and S2, Figures S1 and S2, and Tables S1–S10). This study has been supported by the National Key Research and Development Program of China (2016YFC0202200) and the Guangdong Provincial Project for Innovation and Enterprising (2016ZT06N263).

References

Akimoto, H. (2003). Global air quality and pollution. *Science*, *302*(5651), 1716–1719. https://doi.org/10.1126/science.1092666

Bo, Y., Cai, H., & Xie, S. D. (2008). Spatial and temporal variation of historical anthropogenic NMVOCs emission inventories in China. *Atmospheric Chemistry and Physics*, *8*(23), 7297–7316. https://doi.org/10.5194/acp-8-7297-2008

CAEPI (2015). China development report on desulfurization and denitration industries (in Chinese). *China Environmental Protection Industry*, *12*, 4–23. https://doi.org/10.3969/j.issn.1006-5377.2015.12.001

Cai, H., & Xie, S. D. (2007). Estimation of vehicular emission inventories in China from 1980 to 2005. *Atmospheric Environment*, *41*(39), 8963–8979. https://doi.org/10.1016/j.atmosenv.2007.08.019

Carmichael, G. R., Tang, Y., Kurata, G., Uno, I., Streets, D., Woo, J. H., et al. (2003). Regional-scale chemical transport modeling in support of the analysis of observations obtained during the TRACE-P experiment. *Journal of Geophysical Research*, *108*(D21), 8823. https://doi.org/10.1029/2002JD003117

Carter, W. P. (2008). *Reactivity estimates for selected consumer product compounds*. Riverside, CA: California Air Resources Board. Retrieved from http://www.cert.ucr.edu/~carter/pubs/aminrep.pdf

China Automotive Technology Research Center (1986, 1988, 1991, 1993–2015). *China Automotive Industrial Yearbook*. Beijing, China: Machinery Industry Press.

China Electricity Council (1993–2016). *China Electric Power Yearbook*. Beijing, China: China Electric Power Press.

China Meterological Administration (1986–2016). *China Meteorological Yearbook*. Beijing, China: China Meterological Press.

China Petroleum and Chemical Industry Association (1984–1993). *World Chemical Industry Yearbook*. Beijing, China: China Chemical Information Center.

China Petroleum and Chemical Industry Association (1994–2016). *China Chemical Industry Yearbook*. Beijing, China: China Chemical Information Center.

China Strategic Research Ltd. (1999–2016). *China Market Yearbook*. Beijing, China: Foreign Language Press.

Civil Aviation Administration of China (1994, 2002–2016). *Statistics bulletin of civil aviation airport production*. Beijing, China: Development Planning Department of Civil Aviation Administration of China.

Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., et al. (2016). Forty years of improvements in European air quality: Regional policy-industry interactions with global impacts. *Atmospheric Chemistry and Physics*, *16*(6), 3825–3841. https://doi.org/10.5194/acp-16-3825-2016

Editorial Department of China Agricultural Machinery Industry Yearbook (1987, 1992–2016). *China Agricultural Machinery Industry Yearbook*. Beijing, China: China Machinery Industry Press.

Editorial Department of China Agriculture Yearbook (1980–2014). *China Agriculture Yearbook*. Beijing, China: China Agriculture Press.

Editorial Department of China Building Materials Industry Yearbook (1981–2014). *China Building Materials Industry Yearbook*. Beijing, China: Editorial department of China Building Materials Industry Yearbook.

Editorial Department of China Traffic Yearbook (1986–2014). *China Traffic Yearbook*. Beijing, China: China Communications Press.

Energy Information Administration (2017). International Energy Outlook 2017. from U.S. Energy Information Administration. Retrieved from www.eia.gov/ieo

European Monitoring and Evaluation Programme (2013). *EMEP/EEA air pollutant emission inventory guidebook*. Copenhagen: European Environment Agency. Retrieved from https://doi.org/10.2800/92722

Ge, S., Bai, Z. P., Liu, W. L., Zhu, T., Wang, T. J., Qing, S., & Zhang, J. F. (2001). Boiler briquette coal versus raw coal: Part I—Stack gas emissions. *Journal of the Air & Waste Management Association*, *51*(4), 524–533. https://doi.org/10.1080/10473289.2001.10464293

Hao, J. M., Tian, H. Z., & Lu, Y. Q. (2002). Emission inventories of NO_x from commercial energy consumption in China, 1995–1998. *Environmental Science & Technology*, *36*(4), 552–560. https://doi.org/10.1021/es015601k

IPCC (2013). Summary for policymakers. In T. F. Stocker, et al. (Eds.), *Climate change 2013: The physical science basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (pp. 1–30). Cambridge, UK and New York: Cambridge University Press.

Itahashi, S., Uno, I., Yumimoto, K., Irie, H., Osada, K., Ogata, K., et al. (2012). Interannual variation in the fine-mode MODIS aerosol optical depth and its relationship to the changes in sulfur dioxide emissions in China between 2000 and 2010. *Atmospheric Chemistry and Physics*, *12*(5), 2631–2640. https://doi.org/10.5194/acp-12-2631-2012

Jenkins, B., Turn, S., Williams, R., Goronea, M., & Abd-el-Fattah, H. (1996). *Atmospheric pollutant emission factors from open burning of agricultural and forest biomass by wind tunnel simulations. Volume 2. Results, cereal crop residues. Final report*. Davis, CA: University of California.

Jernelov, A. (1983). Acid rain and sulfur dioxide emissions in China. *Ambio*, *12*(6), 362. Retrieved from http://www.jstor.org/stable/4312971

Kadam, K. L., Forrest, L. H., & Jacobson, W. A. (2000). Rice straw as a lignocellulosic resource: Collection, processing, transportation, and environmental aspects. *Biomass and Bioenergy*, *18*(5), 369–389. https://doi.org/10.1016/S0961-9534(00)00005-2

Kato, N., & Akimoto, H. (1992). Anthropogenic emissions of SO_2 and NO_x in Asia: Emission inventories. *Atmospheric Environment. Part A. General Topics*, *26*(16), 2997–3017. https://doi.org/10.1016/0960-1686(92)90291-R

Klimont, Z., Cofala, J., Xing, J., Wei, W., Zhang, C., Wang, S., et al. (2009). Projections of SO_2, NO_x and carbonaceous aerosols emissions in Asia. *Tellus Series B: Chemical and Physical Meteorology*, *61*(4), 602–617. https://doi.org/10.1111/j.1600-0889.2009.00428.x

Klimont, Z., Streets, D. G., Gupta, S., Cofala, J., Fu, L. X., & Ichikawa, Y. (2002). Anthropogenic emissions of non-methane volatile organic compounds in China. *Atmospheric Environment*, *36*(8), 1309–1322. https://doi.org/10.1016/s1352-2310(01)00529-5

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., et al. (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*(21), 11,019–11,058. https://doi.org/10.5194/acp-13-11019-2013

Lefohn, A. S., Husar, J. D., & Husar, R. B. (1999). Estimating historical anthropogenic global sulfur emission patterns for the period 1850–1990. *Atmospheric Environment*, *33*(21), 3435–3444. https://doi.org/10.1016/s1352-2310(99)00112-0

Lei, Y., Zhang, Q. A., Nielsen, C., & He, K. B. (2011). An inventory of primary air pollutants and CO_2 emissions from cement production in China, 1990–2020. *Atmospheric Environment*, *45*(1), 147–154. https://doi.org/10.1016/j.atmosenv.2010.09.034

Li, C., Yang, X., Li, X., Chen, J., & Qiao, X. (2014). Development of a model for predicting hydroxyl radical reaction rate constants of organic chemicals at different temperatures. *Chemosphere*, *95*, 613–618. https://doi.org/10.1016/j.chemosphere.2013.10.020

Li, G., Bei, N., Cao, J., Wu, J., Long, X., Feng, T., et al. (2017). Widespread and persistent ozone pollution in eastern China during the non-winter season of 2015: Observations and source attributions. *Atmospheric Chemistry and Physics*, *17*(4), 2759–2774. https://doi.org/10.5194/acp-17-2759-2017

Li, L. (2007). The verification of NO_x and SO_2 emission factors in the cement industry in the environmental statistics (in Chinese). *Consultation and Statistics*, *5*, 29–30.

Li, M., Zhang, Q., Kurokawa, J. I., Woo, J. H., He, K., Lu, Z., et al. (2017). MIX: A mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP. *Atmospheric Chemistry and Physics*, *17*(2), 935–963. https://doi.org/10.5194/acp-17-935-2017

Li, M., Zhang, Q., Streets, D. G., He, K. B., Cheng, Y. F., Emmons, L. K., et al. (2014). Mapping Asian anthropogenic emissions of non-methane volatile organic compounds to multiple chemical mechanisms. *Atmospheric Chemistry and Physics*, *14*(11), 5617–5638. https://doi.org/10.5194/acp-14-5617-2014

Li, X. G., Wang, S. X., Duan, L., Hao, J., Li, C., Chen, Y. S., & Yang, L. (2007). Particulate and trace gas emissions from open burning of wheat straw and corn stover in China. *Environmental Science & Technology*, *41*(17), 6052–6058. https://doi.org/10.1021/es0705137

Li, X. Y., & Li, H. P. (2012). Emission and distribution of NH_3 and NO_x in China. *China Environmental Science*, *32*(1), 37–42. https://doi.org/10.3969/j.issn.1000-6923.2012.01.006

Liu, Y., & Zhu, J. (1997). *Practical environmental statistics manual*. Shenyang, China: Liaoning University press.

Lu, Z., Streets, D. G., Zhang, Q., Wang, S., Carmichael, G. R., Cheng, Y. F., et al. (2010). Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. *Atmospheric Chemistry and Physics*, *10*(13), 6311–6331. https://doi.org/10.5194/acp-10-6311-2010

Lu, Z., Zhang, Q., & Streets, D. G. (2011). Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010. *Atmospheric Chemistry and Physics*, *11*(18), 9839–9864. https://doi.org/10.5194/acp-11-9839-2011

MEP (1989–2016). *China environmental status bulletin*. Beijing, China: Ministry of Environmental Protection of China.

MEP (2014a). *China vehicle emission control annual report in 2013*. Beijing, China: Ministry of Environmental Protection of China.

MEP (2014b). *Compilation guide for the air pollutant emission inventory* (in Chinese). Beijing, China: China MEP. Retrieved from http://www.zhb.gov.cn/gkml/hbb/bgg/201501/t20150107_293955.htm

Ministry of Petroleum Industry of China (1959). *Emission standard of petroleum asphalt (SYB 1811-59)* (in Chinese). Beijing, China: Standards Press of China.

Ministry of Transport of the People's Republic of China (2000–2016). *Statistical bulletin of highway and waterway transportation industry*. Beijing, China: Ministry of Transport of the People's Republic of China.

Mo, Z. W., Shao, M., & Lu, S. H. (2016). Compilation of a source profile database for hydrocarbon and OVOC emissions in China. *Atmospheric Environment*, *143*, 209–217. https://doi.org/10.1016/j.atmosenv.2016.08.025

National Bureau of Statistics (1981–2016). *China statistical yearbook*. Beijing, China: China Statistics Press.

National Bureau of Statistics (1985–2016). *China rural statistical yearbook*. Beijing, China: China Statistics Press.

National Bureau of Statistics (1986, 1989, 1991–2016). *China energy statistical yearbook*. Beijing, China: China Statistics Press.

National Bureau of Statistics (1990–2016). *China industrial statistical yearbook*. Beijing, China: China Statistics Press.

National Bureau of Statistics (1994–1984, 1986, 1988–1995, 1998, 2001–2016). *China industry economy statistical yearbook*. Beijing, China: China Statistics Press.

National Petroleum and Chemical Industry Bureau (1992). *Emission standard of petroleum asphalts for road pavement (SH 0522-1992)* (in Chinese). Beijing, China: Standard Press of China.

National Petroleum and Chemical Industry Bureau (2000). *Emission standard of petroleum asphalts for road pavement (SH 0522-2000)* (in Chinese). Beijing, China: Standard Press of China.

National Research Council (2010). *Verifying greenhouse gas emissions: Methods to support international climate agreements* (Vol. 1). Washington, DC: National Academies Press.

National Statistical Bureau (2000). *Compilation of china traffic statistical data (1949–1999)*. Beijing, China: China Statistics Press.

National Statistical Bureau (2010). *China compendium of statistics (1949–2008)*. Beijing, China: China Statistics Press.

National Technical Committee for Standardization of Paint and Pigment (2001). *Indoor decorating and refurbishing materials—Limit of harmful substances of interior architectural coatings (GB18582-2001)* (in Chinese). Beijing, China: Standard Press of China.

National Technical Committee for Standardization of Paint and Pigment (2008). *Indoor decorating and refurbishing materials—Limit of harmful substances of interior architectural coatings (GB18582-2008)* (in Chinese). Beijing, China: Standard Press of China.

National Technical Committee for Standardization of Paint and Pigment (2009). *Limit of harmful substances of exterior wall coatings (GB 24408-2009)* (in Chinese). Beijing, China: Standard Press of China.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., & Hayasaka, T. (2007). An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. *Atmospheric Chemistry and Physics*, *7*(16), 4419–4444. https://doi.org/10.5194/acp-7-4419-2007

Ou, J., Zheng, J., Li, R., Huang, X., Zhong, Z., Zhong, L., & Lin, H. (2015). Speciated OVOC and VOC emission inventories and their implications for reactivity-based ozone control strategy in the Pearl River Delta region, China. *Science of the Total Environment*, *530*, 393–402. https://doi.org/10.1016/j.scitotenv.2015.05.062

Shan, Y., Guan, D., Zheng, H., Ou, J., Li, Y., Meng, J., et al. (2018). China CO_2 emission accounts 1997–2015. *Scientific Data*, *5*, 170201. https://doi.org/10.1038/sdata.2017.201

Shan, Y., Liu, J., Liu, Z., Xu, X., Shao, S., Wang, P., & Guan, D. (2016). New provincial CO_2 emission inventories in China based on apparent energy consumption data and updated emission factors. *Applied Energy*, *184*, 742–750. https://doi.org/10.1016/j.apenergy.2016.03.073

Shi, Y., Xia, Y., Lu, B., Liu, N., Zhang, L., Li, S., & Li, W. (2014). Emission inventory and trends of NO_x for China, 2000–2020. *Journal of Zhejiang University SCIENCE A*, *15*(6), 454–464. https://doi.org/10.1631/jzus.A1300379

Smith, S. J., van Aardenne, J., Klimont, Z., Andres, R. J., Volke, A., & Arias, S. D. (2011). Anthropogenic sulfur dioxide emissions: 1850–2005. *Atmospheric Chemistry and Physics*, *11*(3), 1101–1116. https://doi.org/10.5194/acp-11-1101-2011

Stamenkovic, L. J., Antanasijevic, D. Z., Ristic, M. D., Peric-Grujic, A. A., & Pocajt, V. V. (2016). Estimation of NMVOC emissions using artificial neural networks and economical and sustainability indicators as inputs. *Environmental Science and Pollution Research*, *23*(11), 10,753–10,762. https://doi.org/10.1007/s11356-016-6279-z

Streets, D. G., Bond, T., Carmichael, G., Fernandes, S., Fu, Q., He, D., et al. (2003). An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *Journal of Geophysical Research*, *108*(D21), 8809. https://doi.org/10.1029/2002JD003093

Streets, D. G., Tsai, N. Y., Akimoto, H., & Oka, K. (2000). Sulfur dioxide emissions in Asia in the period 1985–1997. *Atmospheric Environment*, *34*(26), 4413–4424. https://doi.org/10.1016/s1352-2310(00)00187-4

Streets, D. G., Tsai, N. Y., Akimoto, H., & Oka, K. (2001). Trends in emissions of acidifying species in Asia, 1985–1997. *Water, Air, and Soil Pollution*, *130*(1), 187–192. https://doi.org/10.1023/a:1013883628877

Streets, D. G., & Waldhoff, S. T. (1998). Biofuel use in Asia and acidifying emissions. *Energy*, *23*(12), 1029–1042. https://doi.org/10.1016/S0360-5442(98)00033-4

Streets, D. G., & Waldhoff, S. T. (2000). Present and future emissions of air pollutants in China: SO_2, NO_x, and CO. *Atmospheric Environment*, *34*(3), 363–374. https://doi.org/10.1016/s1352-2310(99)00167-3

Streets, D. G., Zhang, Q., Wang, L., He, K., Hao, J., Wu, Y., et al. (2006). Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations. *Journal of Geophysical Research*, *111*, D14306. https://doi.org/10.1029/2006JD007118

Su, S., Li, B., Cui, S., & Tao, S. (2011). Sulfur dioxide emissions from combustion in China: From 1990 to 2007. *Environmental Science & Technology*, *45*(19), 8403–8410. https://doi.org/10.1021/es201656f

Sun, Q., Lu, Y., Fu, L., Tian, H., & Hao, J. (2004). Adjustment on NO_x emission factors and calculation of NO_x emissions in China in the year 2000 (in Chinese). *Techniques & Equipment for Environmental Pollution Control*, *5*(2), 90–94. https://doi.org/10.3969/j.issn.1673-9108.2004.02.023

Tian, H., Zhao, D., & Wang, Y. (2011). Emission inventories of atmospheric pollutants discharged from biomass burning in China (in Chinese). *Acta Scientiae Circumstantiae*, *31*(2), 349–357. https://doi.org/10.13671/j.hjkxxb.2011.02.018

Tian, H. Z. (2003). Studies on present and future emissions of nitrogen oxides and its comprehensive control policies in China, (Doctoral dissertation). Retrieved from (https://tsinghua-primo.hosted.exlibrisgroup.com/primo-explore/fulldisplay?docid=86THU_ALMA_CN21315679540003966&context=L&vid=86THU&search_scope=default_scope&tab=default_tab&lang=zh_CN). Beijing, China: Tsinghua University (in Chinese).

TSU (2001). *Research on air pullution control strategy in Beijing*. Beijing, China.

USEPA (1995). Compilation of Air Pollutant Emission Factors (AP-42). Research Triangle Park, NC: Environmental Protection Agency. Retrieved from https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emission-factors#5thed

USEPA (2007). Emissions factor uncertainty assessment. Research Triangle Park, NC: Environmental Protection Agency. Retrieved from http://www.epa.gov/ttn/chief/efpac/uncertainty.html

Wang, R., Tao, S., Shen, H., Huang, Y., Chen, H., Balkanski, Y., et al. (2014). Trend in global black carbon emissions from 1960 to 2007. *Environmental Science & Technology*, *48*, 6780–6787. https://doi.org/10.1021/es5021422

Wang, S., Wei, W., Du, L., Li, G., & Hao, J. (2009). Characteristics of gaseous pollutants from biofuel-stoves in rural China. *Atmospheric Environment*, *43*(27), 4148–4154.

Wang, S., & Zhang, C. (2008). Spatial and temporal distribution of air pollutant emissions from open burning of crop residues in China (in Chinese). *Sciencepaper online*, *3*(5), 329–333. https://doi.org/10.3969/j.issn.2095-2783.2008.05.005

Wang, T., Xue, L., Brimblecombe, P., Lam, Y. F., Li, L., & Zhang, L. (2017). Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects. *Science of the Total Environment*, *575*, 1582–1596. https://doi.org/10.1016/j.scitotenv.2016.10.081

Wang, W., Wang, W., Zhang, W., & Hong, S. (1996). Geographycal distribution of SO_2 and NO_x emission intensities and trends in China (in Chinese). *China Environmental Science*, *16*(03), 161–167.

Wang, Y., Hao, J., Mcelroy, M. B., & Munger, J. W. (2009). Ozone air quality during the 2008 Beijing Olympics: Effectiveness of emission restrictions. *Atmospheric Chemistry and Physics Discussions*, *9*(2), 5237–5251. https://doi.org/10.5194/acp-9-5237-2009

Wang, Y., Zhang, Q. Q., He, K., Zhang, Q., & Chai, L. (2013). Sulfate-nitrate-ammonium aerosols over China: response to 2000–2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia. *Atmospheric Chemistry and Physics*, *13*(5), 2635–2652. https://doi.org/10.5194/acp-13-2635-2013

Wei, W., Wang, S., Chatani, S., Klimont, Z., Cofala, J., & Hao, J. (2008). Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China. *Atmospheric Environment*, *42*(20), 4976–4988. https://doi.org/10.1016/j.atmosenv.2008.02.044

Wei, W., Wang, S., Hao, J., & Cheng, S. (2011). Projection of anthropogenic volatile organic compounds (VOCs) emissions in China for the period 2010–2020. *Atmospheric Environment*, *45*(38), 6863–6871. https://doi.org/10.1016/j.atmosenv.2011.01.013

Wu, R., Bo, Y., Li, J., Li, L., Li, Y., & Xie, S. (2016). Method to establish the emission inventory of anthropogenic volatile organic compounds in China and its application in the period 2008–2012. *Atmospheric Environment*, *127*, 244–254. https://doi.org/10.1016/j.atmosenv.2015.12.015

Wu, R., & Xie, S. (2017). Spatial distribution of ozone formation in China derived from emissions of speciated volatile organic compounds. *Environmental Science & Technology*, *51*(5), 2574–2583. https://doi.org/10.1021/acs.est.6b03634

Xia, Y., Zhao, Y., & Nielsen, C. P. (2016). Benefits of China's efforts in gaseous pollutant control indicated by the bottom-up emissions and satellite observations 2000–2014. *Atmospheric Environment*, *136*, 43–53. https://doi.org/10.1016/j.atmosenv.2016.04.013

Xue, L. K., Wang, T., Gao, J., Ding, A. J., Zhou, X. H., Blake, D. R., et al. (2014). Ground-level ozone in four Chinese cities: Precursors, regional transport and heterogeneous processes. *Atmospheric Chemistry and Physics*, *14*(23), 13,175–13,188. https://doi.org/10.5194/acp-14-13175-2014

Yang, W. (2016). Present situation, market supply and demand trend of coke industry in China (in Chinese). *Coal Processing and Comprehensive Utilization*, *11*(8), 8–13. https://doi.org/10.16200/j.cnki.11-2627/td.2016.08.002

Yu, S., Yuan, J., & Liang, X. (2017). Trends and spatiotemporal patterns of tropospheric NO_2 over China during 2005–2014. *Water, Air, and Soil Pollution*, *228*(11). https://doi.org/10.1007/s11270-017-3641-9

Yu, T. Y., Lin, C. Y., & Chang, L. F. W. (2012). Estimating air pollutant emission factors from open burning of rice straw by the residual mass method. *Atmospheric Environment*, *54*, 428–438. https://doi.org/10.1016/j.atmosenv.2012.02.038

Yuan, W., Chen, Z., Cao, W., & He, H. (2005). New standard of air pollutant emission in cement industry (in Chinese). *Jiangsu Building Materials*, (2), 16–27.

Yumimoto, K., Uno, I., & Itahashi, S. (2014). Long-term inverse modeling of Chinese CO emission from satellite observations. *Environmental Pollution*, *195*, 308–318. https://doi.org/10.1016/j.envpol.2014.07.026

Zhang, C. (2008). Studies on present and future emissions of PM, SO_2, NO_x, (Master's thesis) (in Chinese). Retrieved from https://tsinghua-primo.hosted.exlibrisgroup.com/primo-explore/fulldisplay?docid=86THU_ALMA_CN21346648230003966&context=L&vid=86THU&search_scope=default_scope&tab=default_tab&lang=zh_CN. Beijing, China: Tsinghua University.

Zhang, C., Wang, S., Xing, J., Zhao, Y., & Hao, J. (2008). Current status and future projections of NO_x emissions from energy related industries in China (in Chinese). *Acta Scientiae Circumstantiae*, (12), 2470–2479. https://doi.org/10.13671/j.hjkxxb.2008.12.026

Zhang, J., Smith, K., Ma, Y., Ye, S., Jiang, F., Qi, W., et al. (2000). Greenhouse gases and other airborne pollutants from household stoves in China: A database for emission factors. *Atmospheric Environment*, *34*(26), 4537–4549.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., et al. (2009). Asian emissions in 2006 for the NASA INTEX-B mission. *Atmospheric Chemistry and Physics*, *9*(14), 5131–5153. https://doi.org/10.5194/acp-9-5131-2009

Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., et al. (2007). NO_x emission trends for China, 1995–2004: The view from the ground and the view from space. *Journal of Geophysical Research*, *112*, D22306. https://doi.org/10.1029/2007JD008684

Zhang, Y., Li, X., Li, M., Zheng, Y., Geng, G., Hong, C., et al. (2018). Reduction in black carbon light absorption due to multi-pollutant emission control during APEC China 2014. *Atmospheric Chemistry and Physics*, 1–24. https://doi.org/10.5194/acp-2018-274

Zhao, B., Wang, S. X., Liu, H., Xu, J. Y., Fu, K., Klimont, Z., et al. (2013). NO_x emissions in China: Historical trends and future perspectives. *Atmospheric Chemistry and Physics*, *13*(19), 9869–9897. https://doi.org/10.5194/acp-13-9869-2013

Zhao, Y., Nielsen, C., Lei, Y., McElroy, M., & Hao, J. (2011). Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China. *Atmospheric Chemistry and Physics*, *11*(5), 2295–2308.

Zhao, Y., Nielsen, C. P., McElroy, M. B., Zhang, L., & Zhang, J. (2012). CO emissions in China: Uncertainties and implications of improved energy efficiency and emission control. *Atmospheric Environment*, *49*, 103–113. https://doi.org/10.1016/j.atmosenv.2011.12.015

Zhao, Y., Zhang, J., & Nielsen, C. (2013). The effects of recent control policies on trends in emissions of anthropogenic atmospheric pollutants and CO_2 in China. *Atmospheric Chemistry and Physics*, *13*(2), 487–508. https://doi.org/10.5194/acp-13-487-2013

Zheng, J., Shao, M., Che, W., Zhang, L., Zhong, L., Zhang, Y., & Streets, D. (2009). Speciated VOC emission inventory and spatial patterns of ozone formation potential in the Pearl River Delta, China. *Environmental Science & Technology*, *43*(22), 8580–8586. https://doi.org/10.1021/es901688e