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Particulate matter pollution over China and the effects of control policies

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29 **Abstract**

30 China is one of the regions with highest PM_{2.5} concentration in the world. In this study, we
31 systematically review the spatio-temporal distribution of PM_{2.5} mass concentration and components in
32 China and the effect of control measures on PM_{2.5} concentrations. Annual averaged PM_{2.5} concentrations
33 in Central-Eastern China reached over 100 µg/m³, in some regions even over 150 µg/m³. In 2013, only
34 4.1% of the cities attained the annual average standard of 35 µg/m³. Aitken mode particles tend to
35 dominate the total particle number concentration. Depending on the location and time of the year, new
36 particle formation (NPF) has been observed to take place between about 10 and 60% of the days. In most
37 locations, NPF was less frequent at high PM mass loadings. The secondary inorganic particles (i.e.
38 Sulfate, Nitrate and Ammonium) ranked the highest fraction among the PM_{2.5} species, followed by
39 organic matters (OM), crustal species and element carbon (EC), which accounted for 6–50%, 15–51%,
40 5–41% and 2–12% of PM_{2.5}, respectively. In response to serious particulate matter pollution, China has
41 taken aggressive steps to improve air quality in the last decade. As a result, the national emissions of
42 primary PM_{2.5}, sulfur dioxide (SO₂), and nitrogen oxides (NO_x) have been decreasing since 2005, 2006,
43 and 2012, respectively. The emission control policies implemented in the last decade could result in
44 noticeable reduction in PM_{2.5} concentrations, contributing to the decreasing PM_{2.5} trends observed in
45 Beijing, Shanghai, and Guangzhou. However, the control policies issued before 2010 are insufficient to
46 improve PM_{2.5} air quality notably in future. An optimal mix of energy-saving and end-of-pipe control
47 measures should be implemented, more ambitious control policies for NMVOC and NH₃ should be
48 enforced, and special control measures in winter should be applied, 40-70% emissions should be cut off
49 to attain PM_{2.5} standard.

50 **Key words:** PM_{2.5}; number concentration; chemical speciation; control measures; China

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75 1. Introduction

76 China is one of the regions with highest concentration of fine particulate matter with aerodynamic
77 diameter equal to or less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) in the world (van Donkelaar et al., 2015). With the rapid
78 growth of economy and urbanization, the air pollution emissions continually increase since 2000. The
79 population-weighted average $\text{PM}_{2.5}$ concentration reached $59 \mu\text{g}/\text{m}^3$ in 2010. More than 80% people lived
80 in the region where air quality did not reach the air quality standard ($35 \mu\text{g}/\text{m}^3$) in 2010 (Apte et al.,
81 2015). The premature mortality caused by $\text{PM}_{2.5}$ is 1.27 million in China (Apte et al., 2015; Lim et al.,
82 2012). Since June 2000, the Ministry of Environmental Protection of China (MEP) started to publish a
83 daily air pollutant index (API), an integrated index calculated from daily concentrations of SO_2 , NO_2 ,
84 and PM_{10} (particulate matter with an aerodynamic diameter of $10 \mu\text{m}$ or less). API data was used to
85 estimate PM_{10} concentrations in 86 Chinese cities and to analyze the long-term variation of PM_{10} across
86 China (Cheng et al., 2013; Qu et al., 2010; Yang, 2009). In 2012, China released a new ambient air quality
87 standard, which was effective in January 1st, 2016, including $\text{PM}_{2.5}$ as a pollutant for the first time (Jiang
88 et al., 2015a). Following that, 74 Chinese cities including Beijing and Shanghai started to monitor hourly
89 concentrations of $\text{PM}_{2.5}$, PM_{10} , SO_2 , NO_2 , CO , and O_3 . Since January 1st, 2013, these data have been
90 available to the public online in real time. By February 2014, the number of cities releasing hourly $\text{PM}_{2.5}$
91 concentrations increased to 190. This dataset offers a good opportunity for us to analyze the spatial
92 distribution and interannual trend of $\text{PM}_{2.5}$ concentration in China.

93 Faced with serious particulate matter pollution, China has taken substantial measures to improve
94 energy efficiency and reduce emissions of air pollutants in the last decade. During the 11th Five Year Plan
95 period (2006-2010), the Chinese government set a target to reduce national SO_2 emissions by 10%. This
96 target was overfilled, such that the emissions of total suspended particulate (TSP) and SO_2 decreased
97 from 2005 to 2010 by 39.0% and 14.3%, respectively (Chinese Environmental Statistical Bulletin,
98 <http://zls.mep.gov.cn/hjtj/>). During the 12th Five Year Plan period (2011–2015), China planned additional
99 10%, and 8% reductions for NO_x emissions and SO_2 emissions, respectively. Inspired by frequent, long-
100 lasting and large-scope heavy haze pollution in eastern China recently, especially in January 2013,
101 Chinese government issued the "Action Plan on Prevention and Control of Air Pollution" in September
102 2013 (hereinafter referred to as Action Plan). It is valuable for both scientific research and decision-
103 making to systematically evaluate the effect of these control measures on national and regional $\text{PM}_{2.5}$
104 pollution. Moreover, given the elevated $\text{PM}_{2.5}$ concentrations in China, current control legislations are
105 still insufficient for the attainment of ambient air quality standard, which requires large and simultaneous
106 reductions of both primary particles and multiple gaseous precursors (Wang and Hao, 2012). To guide
107 future decision-making, we also need to evaluate how future control policies can alter emission trends
108 and $\text{PM}_{2.5}$ concentrations.

109 In this study, we first review the spatial distribution and temporal trend of $\text{PM}_{2.5}$ mass concentration
110 (Section 2), number concentration (Section 3), and its chemical components (Section 4). Then, we review
111 the effect of recent and future control measures on particulate matter pollution in China (Section 5).
112 Finally, we give suggestions for future control activities.

113 2. PM_{2.5} mass concentration across China

114 2.1. Spatial and temporal distribution of PM_{2.5} pollution

115 Due to the limitation of PM_{2.5} observation network, only a few studies observed the spatial
116 distribution of PM_{2.5} pollution. Wang et al. (2015c) reported concentrations of PM₁₀, PM_{2.5} and PM₁
117 monitored at 24 China Atmosphere Watch Network (CAWNET) stations from 2006 to 2014. In CAWNET,
118 PM₁₀, PM_{2.5}, and PM₁ concentrations were monitored by GRIMM EDM 180 environmental dust monitor
119 instruments with 31 different size channels. The instrument is designed to measure the particle number
120 size distribution and particulate mass, based on a light scattering measurement. GRIMM-developed
121 protocols were used to convert the measured number size distribution to a mass concentration consistent
122 with U.S. Environmental Protection Agency protocols for measuring PM using the aerodynamic diameter.
123 Although several studies reported that results from GRIMM were comparable with those from tapered
124 element oscillating microbalance (TEOM) (Grimm and Eatough, 2009; Hansen et al., 2010; Sciare et al.,
125 2007; Zhao et al., 2011), but in China's thereafter PM_{2.5} network observation (especially in the CEPA's
126 network), the main observation instruments used are beta attenuation monitor. Since 2008, U.S.
127 diplomatic missions in Beijing, Shanghai, Guangzhou, Chengdu, and Shenyang have started to monitor
128 PM_{2.5} concentrations using a beta attenuation monitor (BAM-1020, MetOne) and made that data
129 available to the public (see www.stateair.net). The start date, geographic coordinates were shown in Table
130 S1. Martini et al. (2015) analyzed the interannual trend, seasonal variation and diurnal variation of PM_{2.5}
131 using this dataset. The 74 Chinese cities including Beijing and Shanghai started to monitor hourly
132 concentrations of PM_{2.5}, PM₁₀, SO₂, NO₂, CO, and O₃ since January 1st, 2013. PM_{2.5} hourly
133 concentrations were measured either by TEOM or by BAM. Qu et al. (2010) provided a review and
134 analysis of China's air pollution from urban scale to regional scale based on the monthly reports from
135 August 2013 to July 2014.

136 Figure 1 shows the average PM_{2.5} concentration of all sites in large and middle-size cities. As shown
137 in Figure 1, from 2013 to 2015, the general spatial patterns are consistent, with higher PM_{2.5} in eastern
138 China and lower pollution in western part. However, regions with annual PM_{2.5} over 80 µg/m³ (red dots
139 in Figure 1a to 1c) is shrinking, from the whole North China Plain region (NCP) and Shanxi, Shanxi,
140 Sichuan Province in 2013 to mainly the JJJ region in 2015. Figure 2 shows the comparison of China's
141 air quality with that in Europe. The PM_{2.5} concentration from 1996 to 2007 of Europe is reproduced based
142 on Putaud et al. (2010). As shown in Figure 2, the decreasing trend is more clearly seen when focusing
143 on the JJJ, YRD and PRD region. The average PM_{2.5} concentration of JJJ, YRD, PRD decreased from
144 110, 70 and 48µg/m³ to 85, 55 and 34 µg/m³, respectively. Only over half of the cities in PRD reached
145 the air quality standard in 2015, but still have a way to go to reach WHO guideline, and higher from
146 Europe.

147 As shown in Figure 3, the ratio of annual average PM_{2.5} concentrations to PM₁₀ showed a clear
148 increasing trend from northern to southern China, which is also supported by Wang et al. (2015c). The
149 isohyetal line was based on annual precipitation of 160 Chinese meteorological stations during 1971–
150 2000 obtained from National Climate Center of China (reproduced from Ling et al., 2014). Clear gradient
151 from north to south of PM_{2.5}/PM₁₀ are consistent with the isohyetal line. The lowest value (0.40-0.50)

152 were seen in arid region of China, like Urumqi, Yinchuan and Hohhot, which are influenced by dust
153 storm. The ratio in most cities in NCP, like Shijiazhuang, Taiyuan and Zhengzhou, are between 0.50-0.56,
154 where annual precipitation is between 400 mm to 600 mm. The ratio in Northeast China, Beijing and
155 Tianjin are about 0.56-0.66. It indicates that the values were influenced by fugitive dust due to the low
156 precipitation amounts in northern China. The highest ratio was for the middle and lower reaches of YRD
157 and PRD regions, which are characterized by the highest precipitation. The cities with best air quality,
158 like Zhuhai, Haikou and Kunming, show a lower ratio of $PM_{2.5}$ to PM_{10} . This is mainly due to the low
159 $PM_{2.5}$ concentration.

160 In terms of seasonal variations, wintertime $PM_{2.5}$ is always higher than that summertime $PM_{2.5}$ in
161 all Chinese cities, as shown in Figure 4e. China is a major monsoon region. The seasonal variations are
162 largely impacted by the variation of temperature and precipitation. The largest seasonal variations
163 occurred in northern and southern China. The high winter/summer ratio in northeastern China is mainly
164 due to the high winter $PM_{2.5}$ concentration associated with heating from coal combustion. In Heilongjiang,
165 Jilin and Liaoning Province, the low temperature in winter resulted in large demand for indoor heating.
166 Taking Harbin as an example, The average temperature in winter is from -17 to -6 °C. Consequently,
167 the corresponding high emissions resulted in high $PM_{2.5}$ concentration of 136 $\mu\text{g}/\text{m}^3$. In comparison, the
168 summertime $PM_{2.5}$ in Harbin is 33 $\mu\text{g}/\text{m}^3$, which is comparable with the JJJ region (Figure 4b). The high
169 winter-summer ratios in the most south marine China (mainly PRD region), in contrast, is mainly due to
170 its low summer $PM_{2.5}$ level. As precipitation in this region is much more abundant in summer than winter
171 in this region (Taking Guangzhou as an example, the average precipitation from 1971 to 2000 was 736
172 mm in summer while 108 mm in winter), the wet depositions can efficiently remove the $PM_{2.5}$ in summer
173 (Andronache, 2003; Sparmacher et al., 1993). Average summer $PM_{2.5}$ concentration in Guangzhou is
174 only 27 $\mu\text{g}/\text{m}^3$, which is lower than the northern provinces like Fujian, Hunan and Jiangxi. In comparison,
175 winter $PM_{2.5}$ in Guangzhou is usually 62 $\mu\text{g}/\text{m}^3$, which is comparable with other southern provinces
176 (Figure 4d).

177 In terms of spatial distribution, the seasonal variations showed both commonalities and differences.
178 For all the major areas of JJJ, YRD and PRD, the winter-peak and summer-trough pattern is always
179 observed. However, the pollution levels of spring and autumn are not the same. For JJJ and PRD area,
180 $PM_{2.5}$ in spring is usually lower than autumn, while the trend in the YRD is the opposite.

181 The outbreak of the haze episodes also have strong seasonal characteristics. Zheng et al. (2016) and
182 Cheng et al. (2013, 2014) analyzed the episode pattern and caused of Beijing and YRD with year-long
183 observation data, respectively. Severe haze episodes frequently occurred in winter, June, and October for
184 both regions. In spring, the dominant type is mixed haze-dust. The ratio of $PM_{2.5}$ in PM_{10} is minimum
185 among four seasons. The contribution of crustal elements increases. The severe pollution in summer is
186 related to the intensive biomass-burning activities. The contribution of OM and EC increases during the
187 episodes. In fall and winter, the episodes are linked to unfavorable synoptic conditions, like stable
188 Planetary Boundary Layer (PBL) and low wind speed. Regional transport also plays an important role
189 during the heavy pollution episodes (Zheng et al. (2016). The increase of SNA is mainly attributed to the
190 heavy pollution episodes.

191 2.2. PM_{2.5} mass concentration in megacities

192 Figure 5 presents the PM_{2.5} observation studies in three megacities, i.e., Beijing (a), Shanghai (b),
193 and Guangzhou (c). The detail sampling and chemical analysis method are listed in the Tables S2, S3,
194 and S4, respectively. As shown in Figure 5 (Cao et al., 2012; Chen et al., 2014; Dan et al., 2004; Duan et
195 al., 2006; Han et al., 2005; He et al., 2001; Jung et al., 2009; Lin et al., 2009; Schleicher et al., 2010; Shi
196 et al., 2003; Song et al., 2012; Sun et al., 2004; Wang et al., 2009; Wang et al., 2007; Wang et al., 2005;
197 Yang et al., 2011a; Yu et al., 2013; Yu et al., 2014; Zhang et al., 2013; Zhao et al., 2013e; Zhao et al.,
198 2009), the PM_{2.5} mass concentrations decreased from 147 µg/m³ in 2002 to 81 µg/m³ in 2015. All
199 observations far exceed the national standard level 2, i.e. annual average of 35 µg/m³, according to the
200 National Ambient Air Quality Standards (GB3095-2012). Different from ground-based studies, AOD in
201 Beijing shows a slowly increase trend in Beijing (Peng et al., 2016) from 1999 to 2011. It might be caused
202 by several reasons. First, AOD is the column-sum of aerosol extinction. The upper extinction represents
203 more like regional characteristics. During that period, the emission in Hebei Province increases a lot,
204 which impacts the AOD value in Beijing. Second, the mass extinction coefficient might increase due to
205 the change of component and size distribution of PM_{2.5} during 1999 to 2011. Besides, the variation of
206 PBL height might also cause this effect.

207 Similarly to that in Beijing, the PM_{2.5} concentration in Shanghai, as shown in Figure 5b, also showed
208 a generally decreasing trend, from 95 ug/m³ in 2004 to 53 ug/m³ in 2015 (Cao et al., 2013; Du et al.,
209 2011; Feng et al., 2009; Huang et al., 2012a; Kan et al., 2007; Wang et al., 2016; Wang et al., 2015a;
210 Wang et al., 2006b; Xu et al., 2002; Zhang et al., 2015b; Zhao et al., 2015a). Since 2010, the level showed
211 a somewhat stable value around 55 ug/m³, which is about 30% lower than the average level in Beijing.
212 Another interesting feature is that suburban PM_{2.5} level (Jiading District) in Shanghai is generally higher
213 than the urban level. Moreover, this feature is not seasonal-specific, as it was observed in annual average
214 (e.g., in 2006), in winter 2005 as well as in summer 2006 (Feng et al., 2009). This phenomenon can be
215 explained by the impact of regional transport from northern part, or a possible influence from substantial
216 secondary aerosols in downwind of megacities.

217 Not like that in Beijing and Shanghai, PM_{2.5} in Guangzhou, shown as Figure 5c, showed more
218 significant decreasing trend until 2015. The concentration level has fallen from 91 ug/m³ in 2002 to 39
219 ug/m³ in 2015, with a decreasing rate of 57%. In 2015, the annual average PM_{2.5} concentration in
220 Guangzhou reached about 35 ug/m³. The low PM_{2.5} concentration in Guangzhou is caused by several
221 reasons. First, as shown in Figure 5 (c), the precipitation in Guangzhou is much higher than that in Beijing.
222 The plentiful rainfall benefits the removal of air pollution. And the emission intensity in PRD is smaller
223 than JJJ and YRD (Zhao et al., 2013a). Judging from the observation in 2002, Guangzhou also showed
224 the trend of suburban-high and urban-low pattern as that in Shanghai, which might be attributed to the
225 shift in economic and industrial structures over the decade.

226 3. PM number concentration

227 3.1. General characteristics

228 Table 1 summarizes the observed total and size-segregated particle number concentrations for those
229 published data sets that cover at least one year of measurements. We can see that the site-average total
230 particle number concentration varies from about 2,000 cm⁻³ at a remote high-altitude site (Waliguan, 3.8
231 km above sea level) to values in excess of 20,000 cm⁻³ in urban areas. In addition to these data, Peng et
232 al. (2014) reported similar data from 13 sites based on field measurement campaigns that lasted from a
233 few weeks to a couple of months between 2007 and 2011. They found that the average total particle
234 number concentration varied between 13,700 and 28,400 cm⁻³ in urban sites, between 10,200 and 16 600
235 cm⁻³ in regional sites, and between 5,700 and 7,200 cm⁻³ in coastal or background sites. The total particle
236 number concentrations reported in the urban areas of China are comparable to those measured in
237 European urban environments (Hofman et al., 2016; Putaud et al., 2010). In China, total particle number
238 concentrations do not decrease much when moving from urban to rural or even remote environments.
239 This is contrary to Europe where the average total particle number concentrations are typically between
240 about 1,000 and 5,000 cm⁻³ in rural environments and mostly below 1,000 cm⁻³ in rural environments
241 (Asmi et al., 2011; Putaud et al., 2010).

242 Sub-micron particles are usually divided into three modes: the nucleation mode (particles smaller
243 than about 20–30 nm in diameter), Aitken mode (particles from 20–30 nm up to about 100 nm in diameter)
244 and accumulation mode (particles larger than about 100 nm in diameter). In China, Aitken mode particles
245 tend to dominate the total particle number concentration (Table 1, Peng et al., 2014), which is similar to
246 observations made in Europe (Asmi et al., 2011). Exceptions for this feature are the periods of active
247 new particle formation when high concentrations of nucleation mode particles are present, as well as
248 highly-polluted conditions when a big share of the total particle number concentration may be located in
249 the accumulation mode size range (section 4.2).

250 Total particle number concentrations seem to have a relatively weak annual cycle in China, with the
251 reported seasonally-averaged particle number concentrations being typically within a factor two at
252 individual sites (Kivekäs et al., 2009; Qi et al., 2015; Shen et al., 2016b; Shen et al., 2011; Wu et al.,
253 2008; Zhao et al., 2015b). In most locations, the total particle number concentration tend to be the highest
254 in spring. The seasonal cycles of particle concentrations in different size modes do not show any
255 consistent pattern between the different sites, even though the available data are too few for drawing a
256 firm conclusion on whether such a pattern might exist.

257 For all sites in Table 1, the diurnal cycle of the particle number concentration was found to be the
258 strongest for nucleation mode particles and weakest for accumulation mode particles. The strong diurnal
259 cycle of nucleation mode particles is quite expected, as these particles have much shorter atmospheric
260 lifetimes than either Aitken or accumulation mode particles (e.g. Williams et al., 2002), in addition to
261 which their main sources are active mainly during the daytime. The less pronounced diurnal variability
262 of accumulation mode particles compared with Aitken mode particles can be explained by their longer
263 atmospheric lifetime, perhaps combined with less severe influence by local primary particle sources (e.g.
264 Paasonen et al., 2016).

265 To our knowledge, no continuous long-term measured data on particle number concentrations (to
266 be able to perform trend analysis) have been published for China. We are therefore not able to estimate
267 trends in PM number concentrations in China similar to PM mass concentrations (see sections 2.1 and 2.

268 2). However, measurements made during several summers before, and during, the 2008 summer Olympic
269 Games in Beijing showed that the pollutant emission reductions implemented during the Olympics
270 decreased considerably the average particle number concentrations in different size modes (Wang et al.,
271 2013b). This suggests that past changes in air pollutant emissions in Chinese megacities have influenced
272 not only PM mass concentrations, but also particle number concentrations.

273 3.2. Connection with sources and pollution episodes

274 The particle number concentration and size distribution at any location is a net result of primary
275 particle emissions into the atmosphere, formation of new aerosol particles in the atmosphere, and
276 transformation of the aerosol population by various growth and removal processes during their
277 atmospheric transport to the location of interest (e.g. Raes et al., 2000). Based on a very recent emissions
278 inventory (Paasonen et al., 2016), the most important primary particle sources in China was estimated to
279 be power production followed by residential and industrial combustion. Road traffic, while estimated to
280 be the most important primary particle source in Europe, Northern and Southern America, Australia and
281 in large parts of Asia, ranks only the fourth primary particle source in China. In terms of the particle
282 number concentration, more than 70% of the emitted primary particles were estimated to be in the Aitken
283 mode in China (Paasonen et al., 2016).

284 Since the first reported observations by Wehner et al. (2004) in Beijing for more than a decade ago,
285 atmospheric new particle formation (NPF) has been recognized to be a potentially very important source
286 of aerosol particles in China (see Kulmala et al., 2016b; Peng et al., 2014; Shen et al., 2016a; and
287 references therein). Depending on the location and time of the year, NPF has been observed to take place
288 between about 10 and 60% of the days. In most locations, NPF was found to less frequent at high PM
289 mass loadings. However, contrary to theoretical expectations (Kulmala et al., 2016b), NPF has in some
290 cases observed to take place even under extremely polluted conditions in China (Nie et al., 2014; Wang
291 et al., 2013c; Xiao et al., 2015). Reported growth rates of particles formed by atmospheric NPF range
292 typically from a few nm/hour up to about 20 nm/hour in China, which means that these particles tend to
293 grow into the Aitken mode within a few hours. Further growth of these particles to the sizes at which
294 they may act as cloud condensation nuclei (CCN), i.e. larger than about 50-100 nm in diameter (Kerminen
295 et al., 2012), will take from a few hours to a couple of days.

296 Distinguishing between primary particles and particles formed by atmospheric NPF is difficult,
297 especially in urban areas (e.g. Ma and Birmili, 2015). This difficulty is also apparent when looking at the
298 few source apportionment studies made for PM number concentrations in China (e.g. Liu et al., 2016b;
299 Tan et al., 2014). Kulmala et al. (2016a) refined the black carbon tracer method introduced by Rodríguez
300 and Cuevas (2007) for separating secondary particles from primary ones based on atmospheric
301 observations. They applied this method for more than a year of continuous measurements in a sub-urban
302 site in Nanjing, China, and estimated that on average about half of the measured accumulation mode
303 particles and more than 80% of the sub-100 nm particles were formed by atmospheric NPF in this
304 environment. A number of studies have investigated CCN production associated with NPF in China (e.g.
305 Peng et al., 2014; Shen et al., 2016a; Wang et al., 2013a; Wiedensohler et al., 2009). These studies report
306 large enhancements in CCN concentrations after NPF and subsequent particle growth, and suggest that
307 the fraction of CCN originating from NPF may exceed 50% during the seasons with active NPF.

308 Furthermore, there are indications that the contribution of NPF to CCN might be higher for urban sites
309 compared with rural or more remote sites (Peng et al., 2014). Put together, it is evident that both primary
310 and secondary particles need to be considered when studying the environmental, health and climatic
311 effects of atmospheric aerosol particles in China.

312 Formation and evolution of air pollution episodes provide unique cases to investigate how PM
313 number concentrations and size distributions are connected with PM mass concentrations. Based on
314 measurements in Beijing, Guo et al. (2014) showed that clean periods with low PM mass concentrations
315 were often accompanied by strong atmospheric NPF, resulting in high total particle number
316 concentrations with number mean diameters well below 50 nm. During the transition from clean to
317 polluted periods, both PM mass concentration and particle number mean diameter increased considerably,
318 whereas NPF became less frequent and weaker so that the total particle number concentration first
319 decreased and then fluctuated at around a relatively constant level. Liu et al. (2016a) found that the
320 transition from clean to polluted conditions can take place rapidly, within a few hours, or then over a
321 period of several days. They also found that the ratio between accumulation mode and Aitken mode
322 particles was clearly higher during more polluted conditions. This suggests that both newly-formed and
323 primary particles are capable of growing into the accumulation mode during the transition from clean to
324 polluted conditions, highlighting the very dynamic nature of the sub-micron particle population in the
325 urban air of China.

326 **4. PM chemical components and their changes**

327 4.1. PM_{2.5} chemical components and speciation

328 PM₁₀ components and their changes are briefly presented based on long-term observations during
329 2006-2013 in the Atmosphere Watch Network (CAWNET), which covers various regions of China and
330 includes urban, rural and remote sites (Zhang et al., 2015d; Zhang et al., 2012). This section mainly
331 focuses on PM_{2.5} components over China. Based on the recent studies covering at least one-year
332 observation with bulk chemical composition, chemical species in PM_{2.5} measured during 2009–2015 in
333 23 provincial capitals and all the 4 municipalities directly under central government were assembled to
334 perform their speciation reconstruction and compare their spatial and temporal variations. A brief
335 introduction of the locations, sampling and analyses, and PM mass concentrations for all these studies
336 are listed in Table S5.

337 Chan and Yao (2008) have summarized chemical compositions in PM_{2.5} in a critical review about
338 air pollution in China, with a focus on Beijing, Shanghai, Pearl Delta Region (including Guangzhou,
339 Shenzhen and Hong Kong) and immediate vicinities. Later on, Yang et al. (2011b) tried to present a
340 relatively big picture of reconstructed PM_{2.5} speciation composition based on the studies conducted in 16
341 locations across China during 1999–2007. It focused on characterization and comparison of temporal and
342 spatial variations in the PM_{2.5} speciation composition at paired rural/urban sites in several representative
343 megacities.

344 As illustrated in Figure 6, the magnitude and spatial pattern of PM_{2.5} mass and chemical speciation
345 varied noticeably over geographic regions in China. The annual mean concentrations of PM_{2.5} mass and
346 chemical components in the 27 major cities are classified into seven geographic regions over China

347 (Figure 6). In general, those cities in North, Central, East, Southern regions represent relatively developed
348 regions, while Southwest, Northeast and Northwest China represent less developed regions.

349 The magnitude and spatial pattern of PM_{2.5} mass and chemical speciation varied noticeably over
350 geographic regions. North region suffered the most severe PM_{2.5} pollution (159 µg m⁻³), followed by
351 Northwest (136 µg m⁻³) and Central China (122 µg m⁻³), while the South region was barely satisfactory
352 (66.6 µg m⁻³). Annual mean concentrations of PM_{2.5} ranged from 25.0 to 273 µg m⁻³, with the highest
353 concentration in Taiyuan in North China and the lowest in Lhasa in Southwest China. Besides Lhasa,
354 only Haikou in South region met the secondary grade mass concentration limit of the National Ambient
355 Air Quality Standards (NAAQS) in China (35 µg m⁻³). By contrast, there are 11 cities with average PM_{2.5}
356 mass in excess of 100 µg m⁻³, among which 5 of 6 cities lie in North, 3 of 4 cities in Northwest China.

357 Compared to the results by Yang et al. (2011b), PM_{2.5} mass in Chongqing decreased largely from
358 129 µg m⁻³ in 2005 to 73.8 µg m⁻³ in 2012, while the changes were not so evident in Beijing, Shanghai,
359 and Guangzhou during the same period. Compared to annual average concentrations in 2015 released by
360 each city's environmental statement, it is common to see the significant reduction. For examples, the
361 values decreased from 123 to 80.6 µg m⁻³ during 2009–2015 in Beijing, from 76.8 to 39.0 µg m⁻³ during
362 2009–2015 in Guangzhou, and from 73.8 µg m⁻³ to 57.0 µg m⁻³ during 2012–2015 in Chongqing.

363 SNA and OM were the dominant components of PM_{2.5} across China. Their mass percentages (sulfate,
364 nitrate, and ammonium separately except in Haikou) and those of other constituents are diagrammed in
365 Figure 7. OM plus SNA accounted for 35–78% of PM_{2.5} mass in all the cities except those devoid of
366 relevant data (i.e., Hefei, Haikou, Lanzhou), which was lower than that (62–90%) in the previous study
367 (Yang et al., 2011b). SNA constituted 6.0–50% of PM_{2.5} with an average value of 34% across China.
368 SNA mass exhibited a clear decreasing gradient from the North and East to the South and Southwest
369 China: the annual mean SNA concentrations were 50.8, 40.5, 23.0 and 22.3 µg m⁻³ in North, East, South
370 and Southwest, respectively. In fact, these regional distribution features of secondary inorganic aerosol
371 were largely coincident with the intensity of industrial development, urbanization and nitrogen fertilizer
372 applications. The maximum SNA level (84.5 µg m⁻³) and fraction (50%) in PM_{2.5} mass both appeared in
373 Jinan, and the mass concentration was higher than the value during the period of 2006–2007 (68.9 µg m⁻³)
374 (Yang et al., 2011a). In Beijing, the fraction was up to 46% and showed a clear growth as well (Yang
375 et al., 2011a). In contrast, extremely low SNA fractions were observed in Kunming (6.0%) and Lhasa
376 (7.8%). It should still hold that the emission intensities of SO₂ and NO₂ in the developed regions are three
377 to four times their corresponding national averages (Yang et al., 2011b), although the mega cities over
378 China have kept controlling coal consumption and enhancing flue gas desulfurization in recent years.

379 Much higher SO₄²⁻ levels (>30 µg m⁻³) were observed in the industrial cities with high coal
380 consumption intensity in North China, such as Shijiazhuang, Taiyuan and Jinan, especially for the latter.
381 In Jinan, SO₄²⁻ accounted for 30% of PM_{2.5} mass, the highest fraction among all the cities, matching the
382 capital city of Shandong province as the maximum anthropogenic SO₂ emissions on a provincial basis.
383 By contrast, the lowest values were seen in Southwest and South China, where there existed frequent
384 acidic precipitations since 1970's or later. On average, the annual mean SO₄²⁻ concentrations in North
385 China were higher than that in South China by near 10 µg m⁻³. This was partially due to additional use
386 of coal for space heating from late fall through early spring in North China due to cold weather.

387 NO_3^- exhibited a similar spatial distribution with the highest concentrations in North China,
388 followed by Central and East China. The highest concentration was seen in Shijiazhuang, while the
389 lowest concentrations were observed in Kunming and Lhasa. It was evident that both concentrations of
390 NO_3^- and NH_4^+ and their fractions in $\text{PM}_{2.5}$ mass increased constantly in recent years in the cities with
391 data for comparison. Take Beijing for example, NO_3^- fraction was much higher in 2012 (16%) (Zhang et
392 al., 2015a) compared to those during the period of 2000–2009 (6.0–9.0%) while SO_4^{2-} fraction was
393 relatively constant (Yang et al., 2011b; Zhang et al., 2013). As a result, the mass ratio of $[\text{NO}_3^-]/[\text{SO}_4^{2-}]$
394 exhibited noticeably growth, likely indicative of enhanced contribution to fine particle pollution in
395 Beijing from mobile sources over stationary sources. It is worthy of note that this phenomenon has been
396 becoming more and more prominent across China, since the emissions of their gaseous precursors (NO_x
397 and SO_2) is experiencing contrary shifts.

398 The annual mean concentrations of NH_4^+ in North and Central China were about twice those in
399 South and Southwest China, while the values in East and Northwest China were comparable. On the
400 basis of single city, Zhengzhou and Taiyuan had much higher NH_4^+ concentrations than other cities. As
401 particulate NH_4^+ is mainly formed from gaseous NH_3 and acidic species through complicated gas-phase
402 and aqueous-phase reactions, and NH_3 emission is not so clear as NO_x and SO_2 , the reason for its regional
403 difference is unclear.

404 OM accounted for 15–51% of $\text{PM}_{2.5}$ mass, thus was a relatively constant and significant contributor
405 to fine particles across China. It is noted that the annual mean concentrations of OM were remarkably
406 higher in North China than in other regions. Unlike the spatial distribution of SNA, the proportion of OM
407 in all the five cities in Southwest China remained larger than in other regions. It is noteworthy that in
408 Kunming and Lhasa, two plateau cities, OM concentrations were near six-fold larger than SNA
409 concentrations. OM fraction in Lhasa was up to 51%. Along with this highest fraction was the high mass
410 ratio of OC to EC (10.2), likely pointing to predominant formation of secondary organic aerosol through
411 photochemical reaction. This supposition was reasonable, owing to strong solar radiation ($> 7500 \text{ MJ}$
412 m^{-2}) over Tibetan Plateau (Ling et al., 2014). This high OC content and OC/EC ratio, together with low
413 sulfate content implied that frequent biomass burning in this region likely played an important role as
414 well.

415 Crustal material is also a major component of $\text{PM}_{2.5}$ in China. Its contribution rose dramatically in
416 certain areas and during certain periods susceptible to be influenced by dust events or relevant
417 anthropogenic activities (Yang et al., 2011b). Thus there existed a great spatial difference (from 5% to
418 41%) and seasonal variations for its loading and fraction in $\text{PM}_{2.5}$ mass. In Northwest China with
419 relatively less vegetation and more arid and semi-arid deserts, crustal material accounted for 41%, 23%
420 and 25% of $\text{PM}_{2.5}$ mass in Yinchuan, Xi'an and Lanzhou, respectively, whereas in East region the fraction
421 dropped to as low as 5%.

422 Annual mean concentrations of EC presented a similar spatial distribution as that of NO_3^- , which
423 were highest in North China, and ranged from 1.0 (Lhasa) to $9.9 \mu\text{g m}^{-3}$ (Hangzhou) across China. The
424 fraction of EC in $\text{PM}_{2.5}$ ranged from 2% to 12% with an average value of 6%. Compared to the earlier
425 results summarized by Yang et al. (2011b), the annual concentrations of EC were decreased more or less
426 in Beijing, Chongqing, and Shanghai, varying from 8.2 to $6.3 \mu\text{g m}^{-3}$, 6.4 to $4.0 \mu\text{g m}^{-3}$, 6.5 to $1.9 \mu\text{g m}^{-3}$

427 ³, respectively.

428 4.2. Organic species in fine particles

429 Organic matter is an important part of the atmospheric aerosols, usually accounting for 20–50% of
430 PM_{2.5} (Gu et al., 2010). However, due to the huge number of organic compounds with various properties
431 presented in aerosols and analytical difficulties, identified organic compounds typically account for 10%
432 or less of OM mass. The percentage could reach above 60% by adopting more complex analytical
433 procedure (Chang et al., 2009). The identified organic compounds mainly include n-alkanes, polycyclic
434 aromatic hydrocarbons and some polar organic compounds such as organic acids, alcohol, aldehyde,
435 ketone, and monosaccharide. The polar organic compounds contribute about 20–60% of solvent
436 extractable organic compounds (SEOC). These SEOC are of great interesting as they contain useful
437 molecular markers which have been successfully used for source identification and source apportionment
438 (Chang et al., 2009; Feng et al., 2015b; Feng et al., 2012b).

439 Table 2 summarizes the concentrations of some important and ubiquitous SEOC in different areas
440 in China. The total concentration of identified species contributes 3–11% of OC mass. Fatty acid usually
441 had the highest proportion. The concentrations of alkanes, sugars and phthalates also exhibited high
442 levels. No significant temporary changes were observed for alkanes, fatty acid, and PAH in PM_{2.5} in
443 Shanghai. Contrarily, the average concentration of alkanes in PM_{2.5} in Beijing during 2006–2007
444 increased by a factor of 2.1 than that in 2002 whereas that of fatty acids decreased by 36% during this
445 period.

446 Besides the perspective of SEOC, many studies have targeted water soluble organic carbon (WSOC)
447 as well because of its important role on hygroscopicity and cloud condensation nuclei activities (Raes et
448 al., 2010). However, identified species including organic acids and some sugars only accounted for about
449 20% of WSOC. Humic-like substances (HULIS) have a considerable concentration and are of importance
450 in WSOC (Zhang et al., 2010). Their concentrations varied from 1 to 13 $\mu\text{g m}^{-3}$ with their carbon fraction
451 contributing 9–72% of WSOC (Zhang et al., 2010). HULIS contains a high density of quinoid units and
452 carboxylate groups but the specific components of HULIS are still unresolved (Zhu et al., 2012).

453 5. Effect of control measures on particulate matter pollution

454 5.1. Recent control measures for particulate matter pollution

455 The changes of PM_{2.5} concentrations are largely driven by air pollution control policies and
456 measures. Our previous papers (Wang and Hao, 2012; Wang et al., 2014b) have reviewed China's air
457 pollution control policies and measures as by 2012. In this study, we briefly summarize the control
458 policies before 2010 and focus on the control policies and measures issued after 2010.

459 China has a long history of controlling particulate matter pollution. In 2006, China set a target to
460 reduce energy use per unit of Gross Domestic Product (GDP) by 20% and national SO₂ emissions by 10%
461 during the “11th five year plan (2006-2010)”. It turned out that national energy use per unit of GDP, SO₂
462 emissions, and TSP emissions decreased by 19.1%, 14.3%, and 39.0% from 2005 to 2010 (Chinese
463 Environmental Statistical Bulletin, <http://zls.mep.gov.cn/hjtj/>; The State Council of the People's Republic
464 of China, 2011). During the 12th Five-Year Plan period (2011–2015), China planned additional 16%, 10%,
465 and 8% reductions for energy use per unit GDP, NO_x emissions, and SO₂ emissions, respectively (The

466 State Council of the People's Republic of China, 2011). Due to frequent and extensive haze episodes
467 since 2011, the Chinese government has taken the most stringent air pollution control measures in history,
468 and released the "Action Plan on Prevention and Control of Air Pollution" in September 2013 (referred
469 to as "Action Plan") (The State Council of the People's Republic of China, 2013). This plan, for the first
470 time, set clear targets for the improvement of environmental quality, i.e., the PM_{2.5} concentrations should
471 be reduced by 25%, 20%, and 15% in the JJJ region, the YRD region, and the PRD region, respectively,
472 measured in 2017 against the 2012 levels. In all other prefecture-level cities, the PM₁₀ concentrations are
473 required to be reduced by 10% during the same period. The "Action Plan" also set up 10 types of
474 measures, including 35 specific measures to assure the achievement of the environmental goals. The
475 release of "Action Plan" marks the transformation from emission-oriented control policies to air quality-
476 oriented control policies in China. The specific control policies and measures released after 2010 and the
477 implementation status as of 2015 are described as follows.

478 According to the "Action Plan", the share of coal in total energy consumption would be decreased
479 to 65% or less by 2017, and total coal consumption would also decrease during 2012-2017 in the three
480 key metropolitan regions. In 2015, the total coal consumption was 2.75 Gtce, 25% higher than that of
481 2010 but 3.7% lower than that of 2014 (National Bureau of Statistics, 2011, 2016). The share of coal in
482 total energy consumption decreased from 68.0% in 2010 to 64.0% in 2015, attaining the 65% target set
483 in the "Action Plan" two years ahead. Accordingly, the share of clean and renewable energy (including
484 natural gas, nuclear power, hydro power, and wind power) increased from 13.0% in 2010 to 17.9% in
485 2015, due to the promotion of clean energy power and the replacement of coal with clean energy
486 (National Bureau of Statistics, 2011, 2016). Besides, the "Action Plan" demands that more than 70% of
487 raw coal should be prepared and separated before being used by 2017. By the end of 2015, the national
488 capacity for preparation and separation of raw coal reached 2.6 Gt, resulting in a preparation and
489 separation fraction of 65.9%, 15% higher than that of 2010 (China National Coal Association, 2016).

490 Another important measure is eliminating outdated production capacity which has low energy
491 efficiency and high emission rates. The "Action Plan" requires that coal-fired boilers under 10t/h should
492 be phased out by 2017 and those under 20t/h should not be constructed in urban areas (The State Council
493 of the People's Republic of China, 2013). By 2015, the JJJ region has successfully phased out small coal-
494 fired boilers with capacity less than 10 t/h in urban areas, and the YRD region and PRD region have also
495 phased out most of the smaller boilers required to be eliminated. In addition, China has eliminated 44
496 million tons of iron capacity, 86 million tons of steel capacity, 263 million tons of cement capacity, and
497 86 million weight boxes of flat glass capacity during 2013-2015 (Chinese Academy of Engineering,
498 2016).

499 "Yellow Label" vehicles, that is, the pre-China I light duty vehicles and pre-China III heavy duty
500 vehicles, accounted for about 10% of the vehicle population but as much as about 50% of emissions of
501 major air pollutants from on-road vehicles in the end of 2013 (Ministry of Environmental Protection of
502 China, 2014c). During 2013-2015, China has phased out about 15.3 million Yellow Label vehicles
503 (Chinese Academy of Engineering, 2016). The remaining Yellow Label vehicles by the end of 2015 are
504 less than 5.7 million (Clean Air Asia, 2015; Ministry of Environmental Protection of China, 2015a),
505 which are expected to be eliminated by the end of 2017 according to the "Action Plan". Besides, in 2012,

506 the central government released an ambitious plan to develop China's domestic electric vehicle industry
507 and markets, setting a target of accumulated production and sales of EVs up to 5 million by 2020. In
508 2015, China's annual EV sales increased to 331 thousand, accounting for 1.3% of its total vehicle sales
509 and a significant jump compared with the 0.3% sales share in 2014 (Wu et al., 2016).

510 China also made tremendous progress in the installation of flue gas desulfurization and flue gas
511 denitrification facilities. The total capacity of coal-fired power plants equipped with flue gas
512 desulfurization facilities increased from 530 GW (83%) in 2010 to 890 GW (over 99%) in 2015, and the
513 capacity of those equipped with flue gas denitrification facilities increased from 80 GW (12%) to 830
514 GW (92%). During 2011-2015, the capacities of sintering machines equipped with flue gas
515 desulfurization and precalcined cement kilns equipped with flue gas denitrification increased from 29
516 thousand m² (19%) and near zero to 138 thousand m² (88%) and 1600 Mt (92%), respectively (Ministry
517 of Environmental Protection of China, 2016). Except for the emission sources above, all catalytic
518 cracking units of petroleum refineries, non-ferrous metal smelting plants, and coal-fired boilers above
519 20t/h shall install FGD by 2017. The dust-removal facilities of coal-fired boilers and industrial kilns are
520 also required to be upgraded by 2017 (The State Council of the People's Republic of China, 2013).
521 Stringent emission standards for a variety of industries have been rapidly issued since 2010 (Wang et al.,
522 2014b). The "Action Plan" demands that special emission limits be established for 25 key industries; by
523 2015, national emission standards for all these key industries have been developed and released (Guo,
524 2015). In 2015, Chinese government set a new target that all eligible coal-fired power plants should attain
525 an "ultralow" emission standard by 2020, with in-stack concentration limits of 10 mg/m³, 35 mg/m³, and
526 50 mg/m³, for TSP, SO₂, and NO_x, respectively (Ministry of Environmental Protection of China, 2015b).

527 China has periodically tightened emission standards for new vehicles and engines based on the
528 European Union standards since 2000 (Wang et al., 2014b). The China IV standard (consistent with Euro
529 IV) for light-duty vehicles was put into effect in 2011. The China IV standard (consistent with Euro IV)
530 for heavy-duty diesel vehicles was originally planned for implementation in 2010, but it was postponed
531 several times until the final enforcement at the beginning of 2015 (Ministry of Industry and Information
532 Technology of China, 2014). The China V emission standard was adopted by Beijing, Tianjin, Shanghai,
533 and some cities in the PRD region before the end of 2015, and by eleven provincial-level regions in
534 Eastern China in April 2016. The scheduled nationwide implementation is at the beginning of 2017
535 (Clean Air Asia, 2016; Wu et al., 2016). For non-road mobile machine, the China III emission standard
536 is schedule to be put into effect in 2016, following the implementation of the China II standard in 2010
537 (Ministry of Environmental Protection of China, 2014b). China also initiated emission controls for
538 vessels in 2015. "Vessel emission control zones" are required to be set aside in the JJJ region, the YRD
539 region, and the PRD region, and all vessels entering such zones must use low-sulfur fuel (with sulfur
540 content less than or equal to 0.5%) no later than January 2019 (Ministry of Transport of China, 2015).

541 However, NMVOC control policies have been evolving very slowly before 2013. Except for the
542 implementation of vehicle standards which removed NMVOC effectively together with other pollutants,
543 national NMVOC control measures or emission standards were limited to fossil-fuel exploitation and
544 distribution and selected solvent products before 2013 (Wang et al., 2014b). In 2015, China launched a
545 comprehensive control program of NMVOC emissions in petrochemical industry, which targeted to

546 reduce over 30% NMVOC emissions from the 2014 levels by 2017 (by 2015 in key metropolitan regions),
547 (Ministry of Environmental Protection of China, 2014a). New emission standards for several industries
548 (petroleum refining, petroleum chemistry, synthetic resin) and a pollution-discharge fee system were
549 developed to facilitate the control measures.

550 In addition, 80.1% of collectable agricultural residue were used as fertilizer, feed, or transformed to
551 clean energy in 2015, attaining the 80% goal of reducing biomass burning (Information office of the
552 Ministry of Agriculture, 2016).

553 5.2. Effect of recent control measures on air pollutant emissions and PM_{2.5} 554 concentrations

555 5.2.1. Effect of control measures on air pollutant emissions

556 Figure 8 summarizes the recent studies with multiple years (≥ 3) of national emission estimates. In
557 this study, we emphasize on inter-annual emission trends and the effect of control measures; the
558 variability in different emission estimates for a specific year is beyond the focus of this study. In addition,
559 with a focus on the temporal trend in the last decade, we only included studies published in or after 2010.

560 For NO_x emissions, the inter-annual trends estimated by different studies agree fairly well with each
561 other. It can be seen that NO_x emissions were decreasing after 2011 or 2012 owing to the implementation
562 of the 12th Five-Year Plan and the "Action Plan". However, different estimates vary to some extent with
563 respect to the decreasing rate. Xia et al. (2016) developed two parallel emission estimates (PRI and STD)
564 to account for the uncertainty in the penetration and removal efficiency of control technologies. They
565 found that the estimate assuming tighter controls (STD), which derived an 8.9% decline from 2010 to
566 2014, was in better agreement with satellite observations. Tsinghua University (Wang et al., 2014b and
567 updates) also got a similar decline rate of 8.6% during 2010-2014. In addition, the Chinese Academy of
568 Engineering, in cooperation with other institutes, conducted a comprehensive mid-term evaluation of
569 "Action Plan on Prevention and Control of Air Pollution" (Chinese Academy of Engineering, 2016) and
570 estimated that national NO_x emissions decreased by as large as 19% during 2013-2015. Based on the
571 analysis above, we conclude that NO_x emissions are largely reduced during 2011-2015 and the 10%
572 reduction target set in the 12th Five-Year Plan have been overfulfilled. The deployment of NO_x removal
573 equipment in power plants and industrial sector is a major contributor to NO_x emission reduction during
574 this period (Chinese Academy of Engineering, 2016; Xia et al., 2016).

575 All studies except for EDGAR show that SO₂ emissions were increasing until 2006, and then
576 declined ever since. With EDGAR excluded, the decline rates from 2005 to 2010 range 6-15% among
577 the studies reviewed. Zhang et al. (2015c) applied a structural decomposition analysis and showed that
578 the SO₂ reduction during this period mainly resulted from improved technological efficiency, including
579 end-of-pipe abatement efficiency (deployment of flue gas desulfurization facilities) and pollutant
580 generation intensity, while the changes in economic structure did not make noticeable contribution to
581 SO₂ reduction. The SO₂ reduction rate from 2010 to 2014 was estimated at 12%, 2%, and 18% by
582 Tsinghua University (Wang et al., 2014b and updates), the PRI case of Xia et al. (2016) and the STD case
583 of Xia et al. (2016), respectively. Similar to NO_x, Xia et al. (2016) indicated that satellite observations
584 agreed better with the STD case with larger decline rate. Chinese Academy of Engineering (2016)

585 reported a SO₂ decreasing rate as high as 29% for the period of 2013-2015. Therefore, China's SO₂
586 emissions have decreased at a high rate since 2010, far surpassing the 8% reduction target of the 12th
587 Five-Year Plan.

588 As for PM₁₀ and PM_{2.5}, the reviewed studies deviated in terms of the emission trends during 2005-
589 2008. The emission inventories developed by Chinese institutes (MEIC; Wang et al., 2014b; Zhao et al.,
590 2013f) revealed a declining trend and while those compiled as a part of global or regional inventories
591 showed an increasing trend. We believe that PM_{2.5} emissions have decreased during 2005-2010 because
592 (1) Chinese domestic estimates took into account more detailed information of control measures; (2) the
593 official statistics showed a 39% decline in TSP emissions (Chinese Environmental Statistical Bulletin,
594 <http://www.mep.gov.cn/zwgk/hjtj/>); and (3) air quality simulation using the estimate of Wang et al.
595 (2014b) agreed well with the observed trend in ambient PM₁₀ concentrations (Zhao et al., 2013a, b). After
596 2010, the PM₁₀ and PM_{2.5} emissions continued to decrease due to a series of control measures described
597 in Section 5.1 especially the release of stringent industrial emission standards. Chinese Academy of
598 Engineering (2016) showed a decline in PM_{2.5} emissions by 20% during 2013-2015, as a consequence of
599 the "Action Plan".

600 All studies show that China's NMVOC emissions increased constantly during 2000-2014 due to
601 inadequate control measures, though the estimated growth rates differ to some extent. However, the
602 implementation of emission standards for vehicles, fuel distribution, and selected solvent products
603 (Section 5.1) have played a role in slowing down the NMVOC emission increase. In particular, the
604 NMVOC emissions from transportation sector have been decreasing since 2005 (Wang et al., 2014b; Wu
605 et al., 2016).

606 There have been few control measures for NH₃ emissions by the end of 2015. All studies reported
607 overall upward trends in NH₃ emissions in the last decade except for Kang et al. (2016), which showed
608 that NH₃ emissions decreased from 2005 to 2007 and remained roughly constant ever since. This
609 difference arises from different estimation methods, especially the treatment of the types of fertilizer and
610 the corresponding emission factors (Kang et al., 2016), rather than the enforcement of control measures.

611 Having reviewed the national emission trends, we will further investigate the emission trends in key
612 metropolitan regions. Figure 9 illustrates the emission estimates during 2005-2014 for the whole country,
613 the JJJ region, the YRD region, and the PRD region. We only include the estimates by Wang et al. (2014b)
614 and subsequent updates, and Xia et al. (2016), because these are the only two studies that include multi-
615 year emission estimates until 2014. We adopt the STD case reported by Xia et al. (2016) because it agrees
616 better with the satellite observations compared with the PRI case (Xia et al., 2016). We can see from
617 Figure 9 that the emissions in the three key metropolitan regions generally present similar temporal trends
618 to those of the whole country. Specifically, NO_x emissions were increasing until about 2011 followed by
619 a subsequent decline; SO₂ and PM_{2.5} emissions were declining while NMVOC emissions were growing
620 in most of 2005-2014. Furthermore, Figure 9 shows that the emissions in the three key metropolitan
621 regions generally increase slower or decrease faster compared with the national emissions, due largely
622 to more stringent control measures in these key regions. This pattern is revealed by both estimates but it
623 is especially pronounced for the estimate of Wang et al. (2014b) and updates. Averaging the two estimates
624 whenever possible, the change rates of NO_x from 2005 to 2014 are 29%, -8%, 2%, and 9% for the whole

625 country, the JJJ region, the YRD region, and the PRD region, respectively. The corresponding change
626 rates are -26%, -48%, -48%, and -31%, respectively, for SO₂, -17%, -26%, -44%, and -40%, respectively,
627 for PM_{2.5}, and 31%, 23%, 44%, and 7%, respectively, for NMVOC. The emission trends in the key
628 regions are at least partly responsible for the observed declining trends in PM_{2.5} concentrations in Beijing,
629 Shanghai, and Guangdong cities (Section 2.2).

630 5.2.2. Effect of long-term control measures on PM_{2.5} concentrations

631 In this section, we review the studies which evaluated the effect of long-term control measures
632 (relative to temporary control measures) during 2005-2015 on the changes of PM_{2.5} concentrations. The
633 reviewed studies are summarized in Table 3.

634 Wang et al. (2010) evaluated the air quality benefits from SO₂ control measures set in China's 11th
635 Five-Year Plan (2006-2010). They estimated that the successful implementation of the national SO₂
636 control policy would reduce total SO₂ emissions by as much as 21% from the 2005 level (52% from the
637 2010 business-as-usual scenario, short as "BAU"). Consequently, the annual mean concentrations of
638 SO₄²⁻ and PM_{2.5} in Eastern China were estimated to decline by 2-16 μg m⁻³ (12-40%) and 3-15 μg m⁻³ (4-
639 25%), respectively, compared with the 2010 BAU scenario. A subsequent post-evaluation presented by
640 Wang et al. (2014a) reported that the SO₂ reduction ratio during the 11th Five-Year Plan was about 14%.
641 They calculated that the SO₂ reduction decreased ambient SO₄²⁻ concentrations by 8%–10% from the
642 2005 level in Eastern China. These two studies focused on the air quality benefits achievable from SO₂
643 controls, and they both assumed constant emissions for other air pollutants. Zhao et al. (2013a) assessed
644 the environmental effects of changes of all major air pollutants from 2005 to 2010; the influence of inter-
645 annual meteorological variability was ruled out by using the same meteorological fields for 2005 and
646 2010. They concluded that PM_{2.5} concentrations decreased by 2-17 μg m⁻³ in most of the North China
647 Plain, the YRD region and the PRD region, while increasing by 4.5-16 μg m⁻³ in most of the Sichuan
648 Basin and Hubei Province, as an integrated effect of the decrease in SO₂/PM_{2.5} emissions and the increase
649 in NO_x, NMVOC and NH₃ emissions. As an effect solely of emission changes, nitrate concentrations
650 increased across most of Eastern China while sulfate and black carbon concentrations decreased in most
651 of Eastern China, which was consistent with the observational trends described in Section 4.1.

652 Zhao et al. (2013d) used the WRF/CMAQ model to investigate the impact of planned NO_x and SO₂
653 control policies in 12th Five-Year Plan on PM_{2.5} concentrations for the period of 2011-2015. Their
654 simulation revealed that the enforcement of planned control measures could reduce NO_x and SO₂
655 emissions from the 2010 levels by 12.2% and 9.6%, respectively (23% and 19% from the 2015 BAU
656 scenario, respectively). Under the planned NO_x control measures, the annual PM_{2.5} concentration was
657 expected to decline by 1.5-6 μg m⁻³ (1.6-8.5%) in the majority of Eastern China, compared with the 2015
658 BAU scenario. The corresponding PM_{2.5} reduction could be 3-8.3 μg m⁻³ (3.2-13%) with the
659 implementation of joint SO₂/NO_x control measures. Another study using similar method (Wang et al.,
660 2014a) indicated that the NO_x control measures envisaged in the 12th Five-Year Plan would reduce the
661 NO₃⁻ concentration in Eastern China by 3-14% from 2010 to 2015.

662 The "Action Plan" required more aggressive control measures and air quality improvement than
663 those set in the 12th Five-Year Plan. As described in Section 5.2.1, Chinese Academy of Engineering

664 (2016) comprehensively evaluated the effect of the “Action Plan” by the end of 2015 and concluded that
665 national SO₂, NO_x, and PM_{2.5} emissions were reduced by as large as 29%, 19%, and 22% during the
666 period of 2013-2015. The emission reductions accounted for a reduction of about 24% in average PM_{2.5}
667 concentrations in 74 key cities, implying a high probability for the attainment and even overfulfilment of
668 the air quality target by the end of 2017 for most cities. Facility upgrading in key industries, industrial
669 structure adjustment, pollution control of coal-fired boilers, and control of fugitive dust were identified
670 as the most effect groups of control measures, accounting for 31%, 21%, 21%, and 15% of the total PM_{2.5}
671 concentration reductions, respectively. In contrast, meteorological variability played a negligible role in
672 the improvement of PM_{2.5} air quality during 2013- 2015.

673 5.3. Effect of future control measures on air pollutant emissions and PM_{2.5} 674 concentrations

675 5.3.1. Effect of future control measures on air pollutant emissions

676 Many studies have projected future air pollutant emissions considering various control policies.
677 Earlier projections (reported in or before 2008 and based on emissions in or before 2005) generally did
678 not anticipate China’s rapid economic growth and stringent control policies in the last decade (Amann et
679 al., 2008; Cofala et al., 2007; Klimont et al., 2001; Klimont et al., 2002; Ohara et al., 2007; Streets and
680 Waldhoff, 2000; van Aardenne et al., 1999). Therefore, these projections usually deviated greatly from
681 the actual emission trends in the last decade (Wang et al., 2014b). To provide an up-to-date evaluation of
682 future emission trends and effects of control policies, we only review emission projections reported after
683 2008, which are summarized in Figure 10.

684 For NO_x, SO₂, and PM_{2.5}, Figure 10 presents the results from four studies. Tsinghua University
685 (Wang et al., 2014b; Zhao et al., 2013c) projected emissions for 2020 and 2030 based on the emissions
686 of 2010. They designed two energy paths, a BAU scenario assuming current regulations and
687 implementation status, and a new policy (PC) scenario assuming that stringent energy-saving policies
688 would be released and more tightly enforced. They also designed three emission control strategies
689 including a [0] strategy based on current regulations and implementation status, a [1] strategy assuming
690 new policies to be released progressively, and a [2] strategy which assumed full application of maximum
691 technically feasible control technologies. The combination of energy paths and control strategies yielded
692 six emission scenarios (i.e., BAU[0], BAU[1], BAU[2], PC[0], PC[1], and PC[2]). Similarly, Nanjing
693 University (Zhao et al., 2014) developed three energy paths, i.e., a current policy scenario (CPS), a new
694 policy scenario (NPS), and a stringent 450 scenario (450 S), and three emission control strategies, i.e., a
695 base case (BAS), a reference case (or “best guess” case, REF), and a case which assumed recently issued
696 emission standards were met by all the sources (STD). The energy paths and emission control strategies
697 were coupled to make nine emission scenarios. The International Institute for Applied Systems Analysis
698 (IIASA, Cofala et al., 2012) projected pollutant emissions until 2030 based on 2010 emissions and four
699 scenarios envisaging energy-saving measures at different stringency levels, which the current policy
700 scenario and the 450 ppm scenario being the least and most stringent, respectively. Cai et al. (2016)
701 evaluated the effect of the “Action Plan” on emissions in 2017 and 2020 by assuming that the control
702 measures set in the action plan would be effectively enforced as of 2017 and would be progressively

703 strengthened as of 2020.

704 With regard to the effect of energy-saving policies, the implementation of aggressive energy paths
705 were expected to decrease NO_x emissions by 29-35%, SO₂ emissions by 25-36%, and PM_{2.5} emissions
706 by 11-29% from the levels of BAU scenario (or current policy scenario, base scenario) in 2030, according
707 to the projections of Tsinghua University (Wang et al., 2014b; Zhao et al., 2013c), Nanjing University
708 and IIASA. Only Tsinghua University (Wang et al., 2014b; Zhao et al., 2013c) and Nanjing University
709 examined the effects of different emission control strategies on future emission trends as of 2030. Since
710 the assumptions underlying their emission control strategies differ, Tsinghua University reported a larger
711 range of emission trajectories than Nanjing University. The fractional differences between the most and
712 least aggressive emission control strategies (coupled with the same energy path) are 72%, 57%, and 69%
713 for NO_x, SO₂, and PM_{2.5} emissions in 2030, respectively, according to the projection of Tsinghua
714 University. The corresponding fractional differences are 50%, 44%, and 31%, respectively, for Nanjing
715 University. Cai et al. (2016) just projected air pollutant emissions as of 2020 but it is the only study that
716 considers the “Action Plan” issued in 2013. Figure 10 illustrates that the change rates of NO_x, SO₂, and
717 PM_{2.5} emissions during 2010-2020 predicted by Cai et al. (2016) resemble the most aggressive scenario
718 of Nanjing University and the second most aggressive scenario (PC[1]) of Tsinghua University (Wang et
719 al., 2014b; Zhao et al., 2013c), implying the effectiveness of the “Action Plan” in reducing air pollutant
720 emissions if enforced successfully.

721 For NMVOC, both Tsinghua University (Wang et al., 2014b; Zhao et al., 2013c) and Wei et al. (2011)
722 showed that NMVOC emission would continue to increase under current legislations. It is promising to
723 achieve noticeable emission reduction by 2020 by implementing new control policies. For the year 2020,
724 the PC[1] scenario of Tsinghua University (Wang et al., 2014b; Zhao et al., 2013c), the advanced control
725 measure scenario of Wei et al. (2011) and the “Action Plan” scenario of Cai et al. (2016) all projected a
726 22-24% reduction in NMVOC emissions from the 2020 BAU (or current legislation) scenario. Tsinghua
727 University (Wang et al., 2014b; Zhao et al., 2013c) was the only study projecting emissions as of 2030,
728 which illustrated a large reduction potential as much as 64% from the BAU projection with the
729 enforcement of the most aggressive control measures.

730 5.3.2. Effect of future control measures on PM_{2.5} concentrations

731 As described in Section 5.1, the “Action Plan” has played a vital role in China’s recent air pollution
732 control activities, and will continue to function until the end of 2017. Cai et al. (2016) evaluated the effect
733 of the “Action Plan” on PM_{2.5} concentration in the JJJ region, the most polluted metropolitan region in
734 China, using the WRF/CMAQ model. They found that the implementation of this plan would lead to
735 larger reduction ratios in the emissions of SO₂, NO_x, PM_{2.5}, and NMVOC (36%, 31%, 30%, and 12%,
736 respectively) in the JJJ region compared with the national average level. Consequently, the ambient
737 annual PM_{2.5} concentration in the JJJ region in 2017 was projected to be 28% lower than those in 2012.
738 With the assumption that the control measures would continue to be strengthened progressively during
739 2018-2020, the annual PM_{2.5} concentration in 2020 could be about 38% lower than that of 2012. Using a
740 similar method, Jiang et al. (2015b) assessed the air quality improvement achievable under the “Action
741 Plan” in the PRD region from 2012 to 2017. Their results showed that the “Action Plan” could lead to
742 effective emission reductions of 34% of SO₂, 28% of NO_x, 26% of PM_{2.5}, and 10% of VOCs. These

743 emission abatements would lower the PM_{2.5} concentration by 17%, thereby attaining the 15% target
744 established in the action plan. It should be noted that Jiang et al. (2015b) assumed that the anthropogenic
745 emissions outside of Guangdong Province would remain constant during the study period. Therefore,
746 larger reduction in PM_{2.5} concentration might be achieved considering the synergistic control outside
747 Guangdong Province.

748 An earlier study by Xing et al. (2011) projected the PM_{2.5} concentrations in 2020 with four scenarios
749 assuming control policies with different levels of stringency using 2005 as the base year. Their reference
750 scenario (the REF[0] scenario) was projected to increase PM_{2.5} concentrations slightly and their most
751 stringent scenario (the PC[2] scenario) was estimated to reduce the average PM_{2.5} concentration in
752 Eastern China from the 2005 level by 16%, slightly less reduction than those of Cai et al. (2016) and
753 Jiang et al. (2015b). Note that even this most stringent scenario of Xing et al. (2011) did not anticipate
754 the recent stringent control policies released after 2007.

755 For the mid-term future (as of 2030), Amann et al. (2008), Madaniyazi et al. (2015), and GBD
756 MAPS Working Group (2016) projected the PM_{2.5} concentrations under various emission scenarios.
757 Amann et al. (2008) used a source-receptor relationship derived from the TM5 model to compute the
758 changes of PM_{2.5} concentrations in response to emission changes. They found that the PM_{2.5} concentration
759 would roughly remain the 2005 level under the baseline scenario assuming that existing policies on
760 energy and environment will be continued and implemented. With the uniform application of advanced
761 emission control technologies, the population-weighted PM_{2.5} concentrations would decline by 44%,
762 from about 80 µg/m³ in 2005 to 45 µg/m³ in 2030. Similarly, the simulation conducted by Madaniyazi et
763 al. (2015) using a global chemical transport model (MIROC-ESM-CHEM) revealed that PM_{2.5}
764 concentration in Eastern China would decrease by 0.62 µg/m³ under a “current legislation” scenario (CLE)
765 and 20.41 µg/m³ under a “maximum technically feasible reduction” scenario (MFR) between 2005 and
766 2030, respectively. Note that these two projections were based on emission scenarios developed before
767 2008, which did not anticipate China’s rapid economic growth and stringent control policies in the last
768 decade and could affect the projections in both baseline and policy scenarios.

769 Recently, GBD MAPS Working Group (2016) projected the PM_{2.5} concentrations in 2030 using the
770 emission scenarios developed by Tsinghua University (Wang et al., 2014b; Zhao et al., 2013c) (see
771 Section 5.3.1 and Figure 10) and subsequently updated based on latest emission estimates in 2011-2013.
772 The results revealed that PM_{2.5} concentrations would decrease by 2.0-5.1 µg/m³ (5-8%) over four major
773 metropolitan regions (the JJJ region, the YRD region, the PRD region, and the SCB region) from 2013
774 to 2030 under the BAU[1] scenario assuming current energy policies and progressively strengthened
775 emission control strategies. Under the PC[2] scenario assuming very stringent energy policies and
776 emission control strategies, the corresponding PM_{2.5} reduction was estimated to be as high as 14.6-40.3
777 µg/m³ (40-52%). Note that NH₃ emissions were assumed to remain unchanged in all 2030 scenarios.
778 Further PM_{2.5} reductions beyond the PC[2] scenario would be expected if NH₃ emissions were
779 significantly reduced.

780 Wang et al. (2015b) investigated the possible pathways for the attainment of China’s new ambient
781 PM_{2.5} standard (35 µg/m³ for annual average concentration). They found that the national emissions of
782 SO₂, NO_x, PM_{2.5}, and NMVOC should be reduced by at least 51%, 64%, 53%, and 36%, respectively,

783 by 2030 from the 2012 levels, and NH₃ emissions should also be reduced slightly. More aggressive
784 control measures are required to be enforced in polluted metropolitan regions. For example, in the JJJ
785 region, emissions of SO₂, NO_x, PM_{2.5}, NMVOC, and NH₃ need to be cut off by at least 59%, 71%, 70%,
786 45%, and 21%, respectively.

787 To sum up the future PM_{2.5} projections reviewed above, we conclude that the control policies issued
788 before 2010 are insufficient for notable improvement of PM_{2.5} air quality. Effective implementation of
789 the “Action Plan” could achieve the planned PM_{2.5} reduction target, and further control policies in the
790 mid-term (as of 2030) are needed to achieve larger PM_{2.5} reductions. Due to elevated PM_{2.5} concentrations
791 nowadays, the attainment of ambient PM_{2.5} standard would require dramatic reduction of primary PM_{2.5}
792 and multiple gaseous precursors.

793 **6. Conclusions and outlook**

794 This review summarized the spatio-temporal distribution of PM_{2.5} pollution in China and reviewed
795 the effects of control measures on PM_{2.5} concentrations.

796 The annual averaged PM_{2.5} concentrations in Central-Eastern China were over 100 µg/m³, in some
797 regions even over 150 µg/m³. In 2013, only 4.1% of the cities attained the annual average standard of 35
798 µg/m³ and much higher than that in Europe. Aitken mode particles tend to dominate the total particle
799 number concentration. Depending on the location and time of the year, NPF has been observed to take
800 place between about 10 and 60% of the days. SNA ranked the highest fraction among the PM_{2.5} species,
801 followed by OM, crustal species and EC, which accounted for 6–50%, 15–51%, 5–41% and 2–12%,
802 respectively.

803 In response to serious particulate matter pollution, China has issued a number of aggressive national
804 work plans to improve air quality in the last decade. Owing to the effective implementation of control
805 measures, the national emissions of primary PM_{2.5}, SO₂, and NO_x have been decreasing since 2005, 2006,
806 and 2011/2012, respectively. Modeling studies showed that the emission control policies implemented in
807 the last decade could result in noticeable reduction in PM_{2.5} concentrations, contributing to the decreasing
808 trends in observed PM_{2.5} concentrations in three megacities (Beijing, Shanghai, and Guangzhou). Further
809 control policies in the mid-term (as of 2030) can potentially lead to larger reduction in PM_{2.5}
810 concentrations. Energy-saving measures and end-of-pipe emission control strategies both have large
811 potentials to reduce air pollutant emissions, therefore they should be applied jointly.

812 We provide the following recommendations according to the review. First, the observation sites for
813 PM_{2.5} mass concentration have covered major cities of China, but PM_{2.5} chemical components were only
814 measured by different institutes with different methods. A PM_{2.5} component observational network
815 should be established to achieve long-term component measurements with unified methods, which
816 benefits the characteristic analysis and source apportionment of PM_{2.5} pollution. Second, the release of
817 “Action Plan” marks the transformation from emission-oriented control policies to air quality-oriented
818 control policies and benefits the improvement of PM_{2.5} air quality in the short term, but no environmental
819 targets or policies have been released beyond 2017. We recommend the government develop a mid-term
820 (as of 2030) ambient PM_{2.5} target to guide China’s future emission control activities. In accordance with
821 the air quality target, provincial and stepped emission reduction targets should be formulated and

822 optimized to assure the achievement of the air quality target. Third, while the emissions of PM_{2.5}, SO₂,
823 and NO_x have started to decline, the emissions of NMVOC and NH₃ have kept increasing or at least
824 remained stable. More ambitious control policies for NMVOC and NH₃ should be gradually enforced to
825 achieve substantial improvement of PM_{2.5} air quality. Fourth, average PM_{2.5} concentrations in winter have
826 been remarkably higher than those in summer, and heavy pollution episodes have frequently occurred in
827 winter. Therefore, special control measures are recommended to be applied in winter, e.g., stringent
828 control of heating boilers, prohibition of small stoves, traffic restriction, suspension of some industrial
829 production, etc. Last but not the least, besides the challenge to mitigate particulate matter pollution, China
830 has committed that national CO₂ emissions should peak before 2030. In this study we showed that both
831 energy-saving measures and end-of-pipe control measures have large emission reduction potentials as of
832 2030, while the latter have played a dominant role in the China's historical emission reductions. We
833 suggest that an optimal mix of energy-saving and end-of-pipe control measures should be implemented
834 to maximize the co-benefits between PM_{2.5} pollution control and climate change mitigation.

835

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1215 Table 1 Summary of measured particle number concentrations in China based on results published in
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 1217 numbers refer to the mean concentration measured at each site and given size range.

Site Description	Study period	Average particle number concentrations (cm ⁻³) in different size ranges, and the total concentration				Reference
Beijing, urban	2004.03–2006.02	3–20 nm 9 000	20–100 nm 15 900	100–1000 nm 7 800	Total 32 800	Wu et al. (2008)
Lanzhou, urban	2012.09–2013.08	10–25 nm 1 800	25–100 nm 16 100	100–1000 nm 5 000	Total 22 800	Zhao et al. (2015b)
Nanjing, sub-urban	2011.12–2013.11	6–30 nm 5 300	30–100 nm 8 000	100–800 nm 5 800	Total 19 200	Qi et al. (2015)
Shandianzi, rural	2008.03–2009.08	3–25 nm 3 600	25–100 nm 4 400	100–1000 nm 3 500	Total 11 500	Shen et al. (2011)
Waliguan, rural, mountain	2005.08–2007.05	12–21 nm 570	21–95 nm 1 100	95–570 nm 430	Total 2000	Kivekäs et al. (2009)
Mt. Tai rural, mountain	2010.07–2012.02	3–25 nm 3 200	25–100 nm 5 200	100–1000 nm 3 400	Total 11 800	Shen et al. (2016b)

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Table 2 Concentrations of SEOs in China

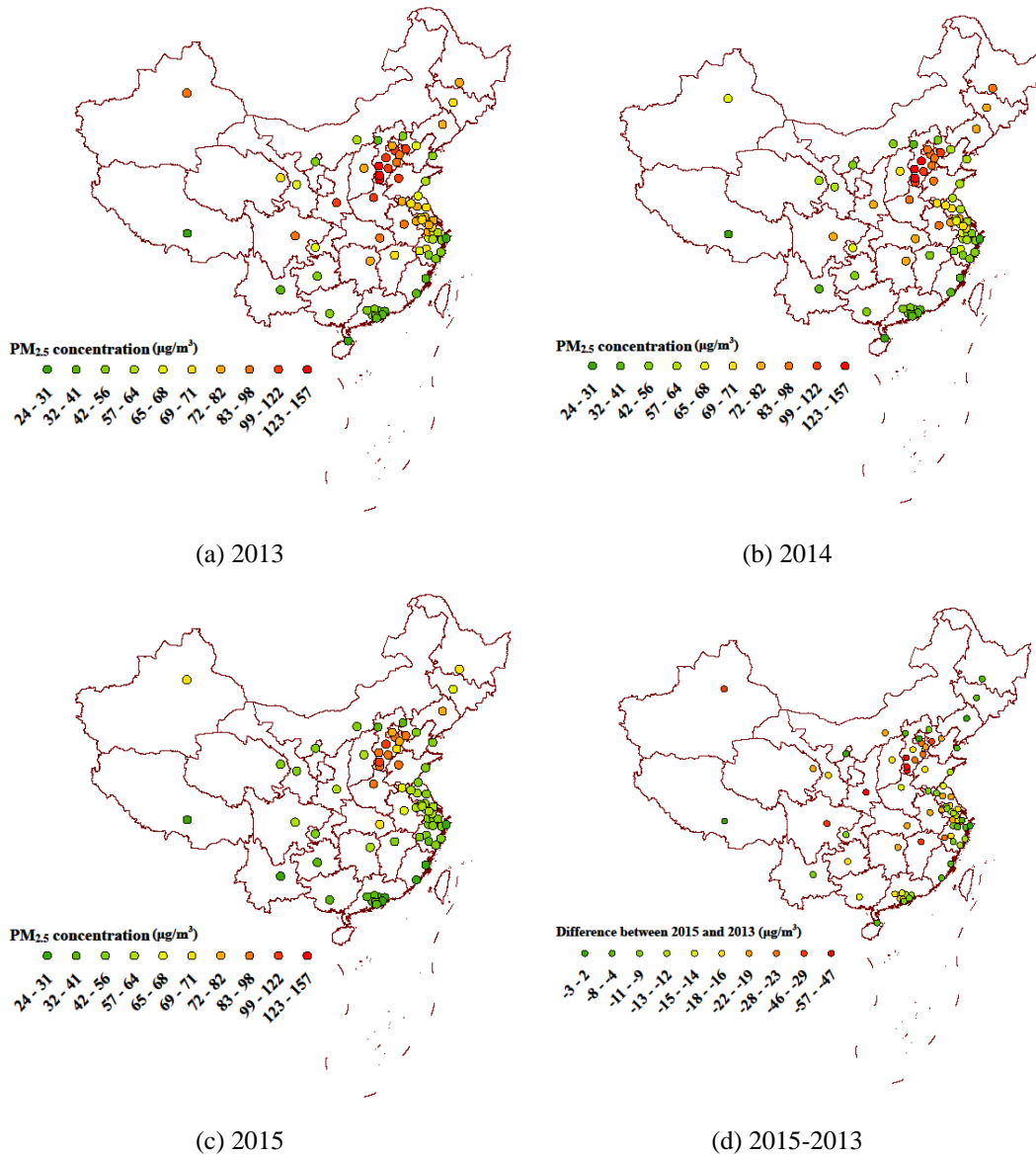
Site-description	Study period	Concentration ($\mu\text{g m}^{-3}$)					Concentration (ng m^{-3})								Reference	
		PM _{2.5}	OC	WSOC	alkanes	fatty acid	sugars	phthalates	fatty alcohols	polyols & polyacids	lignin & resin products	sterols	PAHs	hopanes		
Beijing	Urban	2006.9-2007.8				832	324							113		Li et al. (2013)
Beijing	Capital of China	2003.1, 2003.6-7	89	17.5		268	443	122	290	15	60	10.1	46.3	108	11.2	Wang et al. (2006a)
Beijing	Urban	2002.7, 2002.11		23.5	6.32	322	664							122		Feng et al. (2005); Feng et al. (2006b)
Beijing	Urban	2001.8-2001.7				163	385				26.5			66.2	6.9	Huang et al. (2006)
Changchun	continental & industrial	2003.1, 2003.6-7	97	25.5		353	598	309	264	29.5	132	24.5	114	111	12.4	Wang et al. (2006a)
Changdao island		2003.3-2004.1		8.35	4.08	96.2	174	66						45.1	11.3	Feng et al. (2007)
Chongqing	continental & industrial	2003.1, 2003.6-7	277	67		638	2041	1767	1268	240	302	99	399	361	22	Wang et al. (2006a)
Guangzhou	industrial & commercial	2003.1, 2003.6-7	156	47		575	1859	514	382	46.6	133	100	464	167	30.9	Wang et al. (2006a)
Guangzhou	Urban	2002.12, 2003.7		20.5	6.7	121	740							61.8		Feng et al. (2006b)
Hangzhou	continental	2003.1, 2003.6-7	111	20		250	606	171	390	16	85	14.2	78.5	26.8	4.13	Wang et al. (2006a)
HongKong	coastal & commercial	2003.1, 2003.6-7	63	13		103	274	78.7	288	17	73	3.3	17.8	7.85	2.36	Wang et al. (2006a)
HongKong	Urban	2000.11-2001.11		8.45		57.9	175							8.05		Sin et al. (2005)
Jinchang	Asian dust source regions	2003.1, 2003.6-7	124	15.5		291	370	106	188	31.5	74	8.65	39.6	109	3.75	Wang et al. (2006a)
Lin'an	Background	2008.4-2009-1	59.7	10.3	5.31	60	135	136						25	2.9	Feng et al. (2015a)
Mt. Tai		2006.6		14.5	9.2	117	204	409	255	129	97	6.05	13.5	12.5	1.4	Fu et al. (2012)
Qingdao	coastal	2003.1, 2003.6-7	65.5	11		168	361	89	134	11	64	28	18.5	45.6	3.57	Wang et al. (2006a)
Qingdao		2001.6-2002.5				217	654							87.5		Guo et al. (2003)
Shanghai	Urban	2009.1.21-2.6	91	11		104	210							36.6	6.7	Feng et al. (2012a)
Shanghai	industrial & commercial	2003.1, 2003.6-7	81	15.5		194	463	258	477	26	107	17.3	48	46	9.1	Wang et al. (2006a)
Shanghai	Urban	2002.11, 2003.8		10.3	4.07	83	255							30		Feng et al. (2006a); Feng et al. (2006b)

Site-description	Study period	concentration ($\mu\text{g m}^{-3}$)					concentration (ng m^{-3})								Reference	
		PM _{2.5}	OC	WSOC	alkanes	fatty acid	sugars	phthalates	fatty alcohols	polyols & polyacids	lignin & resin products	sterols	PAHs	hopanes		
Shenzhen	Urban	2009.1-2009.12				56	253					25.2		148	2.51	Huang et al. (2012b)
Taizhou	e-waste	2006.6, 2007.1	87.5	33.1		232	332	179	212					103	11.2	Gu et al. (2010)
Tianjin	industrial	2003.1, 2003.6-7	101	19		225	673	165	541	17.1	101	9.25	43	77	8.4	Wang et al. (2006a)
Wuhan	industrial & commercial	2003.1, 2003.6-7	86	21		219	628	340	303	17	121	23.7	56	60.4	1.8	Wang et al. (2006a)
Xi'an	continental & industrial	2003.1, 2003.6-7	215	60		865	1663	1711	554	286	250	174	736	403	31.9	Wang et al. (2006a)
Xiamen	coastal & commercial	2003.1, 2003.6-7	47.5	12.2		242	300	79.5	175	9.85	83	6.75	10.5	14.3	1.7	Wang et al. (2006a)
Yulin	continental, close to a desert	2003.1, 2003.6-7	85.5	20		372	487	52	162	23.2	89	5.6	283	103	11	Wang et al. (2006a)

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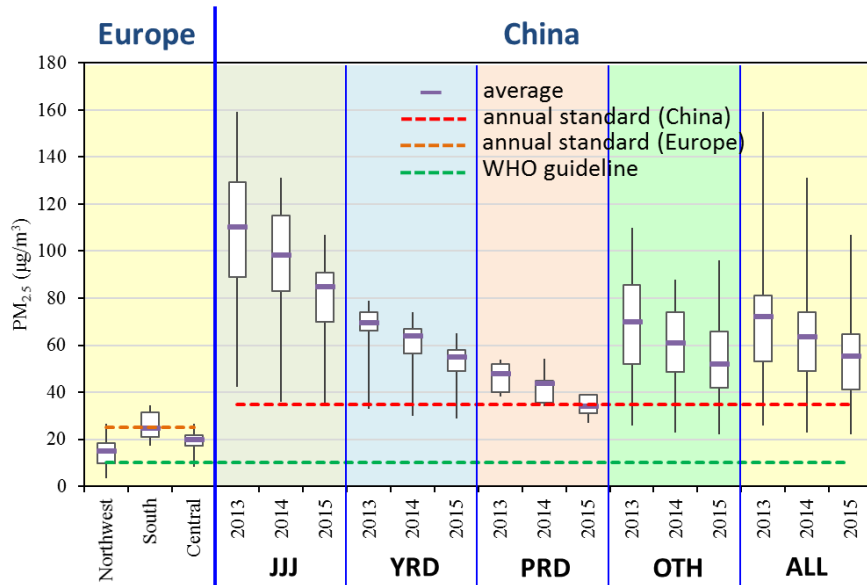
Table 3 Summary of modeling studies evaluating the effect of recent and future control measures on PM_{2.5} concentrations in China

Source	Period	Region	Model	Scenarios	Major results
Effect of recent control measures on PM_{2.5} concentrations					
Wang et al. (2010)	2005-2010: projection	China	MM5/CMAQ	A 2010 BAU scenario, a 2010 SO ₂ control scenario based on the 11 th Five-Year Plan, and a 2010 NO _x control scenario	Under the 2010 SO ₂ control scenario, annual concentrations of SO ₄ ²⁻ and PM _{2.5} in Eastern China were estimated to decline by 2-16 μg m ⁻³ (12-40%) and 3-15 μg m ⁻³ (4-25%), respectively, compared with the 2010 BAU scenario. Under the 2010 NO _x control scenario, NO ₃ ⁻ and PM _{2.5} concentrations decline by 2-7 μg m ⁻³ (20-40%) and 2-14 μg m ⁻³ (3-12%), respectively.
Wang et al. (2014a)	2005-2010: post-evaluation 2010-2015: projection	China	WRF/CMAQ	A 2010 BAU scenario, a 2010 scenario assuming SO ₂ control in the 11 th Five-Year Plan, a 2015 BAU scenario, and a 2015 scenario assuming NO _x control in the 12 th Five-Year Plan	The SO ₂ control in the 11 th Five-Year Plan decreased ambient SO ₄ ²⁻ concentrations by 8%–10% from the 2005 level in Eastern China. NO _x control measures assumed in the 12 th Five-Year Plan would reduce the NO ₃ ⁻ concentration in Eastern China by 3-14% from 2010 to 2015.
Zhao et al. (2013a)	2005-2010: post-evaluation	China	WRF/CMAQ	A 2005 scenario, a 2010 scenario, and a 2005 scenario using the meteorology data of 2010	As an effect solely of emission changes from 2005 to 2010, PM _{2.5} concentrations decreased by 2-17 μg m ⁻³ in most of the North China Plain, the YRD region and the PRD region, while increasing by 4.5-16 μg m ⁻³ in most of the Sichuan Basin and Hubei Province.
Zhao et al. (2013d)	2010-2015: projection	China	WRF/CMAQ	A 2015 BAU scenario, a 2015 NO _x control scenario, and a 2015 joint SO ₂ /NO _x control scenario, both based on 12 th Five-Year Plan	Under the 2015 NO _x control scenario, the annual PM _{2.5} concentration was expected to decline by 1.5-6 μg m ⁻³ (1.6-8.5%) in most of Eastern China, compared with the 2015 BAU scenario. The corresponding PM _{2.5} reduction could be 3-8.3 μg m ⁻³ (3.2-13%) under the joint SO ₂ /NO _x controls.
Chinese Academy of Engineering (2016)	2013-2015: post-evaluation	China	WRF/CMAQ	A 2013 scenario and a 2015 scenario with the effect of “Action Plan” considered	The implementation of “Action Plan” decreased SO ₂ , NO _x , and PM _{2.5} emissions by 29%, 19%, and 22% during 2013-2015, accounting for about 24% reduction in average PM _{2.5} concentrations in 74 key cities. Meteorological variability played a negligible role in the changes of PM _{2.5} concentrations from 2013 to 2015.
Effect of future control measures on PM_{2.5} concentrations					
Xing et al. (2011)	2005-2020	Eastern China	MM5/CMAQ	Four 2020 scenarios assuming different combinations of energy paths and emission control strategies	The reference scenario was projected to increase average PM _{2.5} concentrations in Eastern China by 8% from the 2005 level and the most stringent scenario was estimated to reduce average PM _{2.5} concentration by 16% from the 2005 level.
Cai et al. (2016)	2012-2017	JJJ region	WRF/CMAQ	A 2017 scenario assuming the enforcement of “Action Plan” and a 2020 scenario assuming extrapolation of “Action Plan”	With the implementation of the “Action Plan”, the ambient annual PM _{2.5} concentrations in the JJJ region in 2017 and 2020 were projected to be 28% and 38% lower than those in 2012, respectively.
Jiang et al. (2015b)	2012-2017	PRD region	WRF/CMAQ	A 2017 scenario assuming the enforcement of “Action Plan”	The implementation of the “Action Plan” was estimated to lower the PM _{2.5} concentration in the PRD region by 17% from the 2012 level.
Amann et al. (2008)	2005-2030	China	source-receptor relationship	A 2030 baseline scenario and a 2030 control scenario assuming uniform application of advanced emission control technologies	The PM _{2.5} concentration would roughly remain the 2005 level under the 2030 baseline scenario. In the control scenario, population-weighted PM _{2.5} concentrations would decline by 44% from about 80 μg/m ³ in 2005 to 45 μg/m ³ in 2030.
Madaniyazi et al. (2015)	2005-2030	Eastern China	MIROC-ESM-CHEM	A 2030 “current legislation” scenario (CLE) and a 2030 “maximum technically feasible reduction” scenario (MFR)	PM _{2.5} concentration in Eastern China would decrease by 0.62 μg/m ³ under the CLE scenario and 20.41 μg/m ³ under the MFR scenario by 2030 from the 2005 level.
GBD MAPS Working Group (2016)	2013-2030	China	GEOS-Chem	Four 2030 scenarios assuming different combinations of energy paths and emission control strategies	PM _{2.5} concentrations would decrease by 2.0-5.1 μg/m ³ (5-8%) and 14.6-40.3 μg/m ³ (40-52%) over four major metropolitan regions in China from 2013 to 2030 under the BAU[1] scenario assuming current energy policies and progressively strengthened emission control strategies, and the PC[2] scenario assuming very stringent energy policies and emission control strategies, respectively.
Wang et al. (2015b)	2012-2030	China	CMAQ/RSM	A 2030 scenario designed to meet the ambient PM _{2.5} standard of 35 μg/m ³	National emissions of SO ₂ , NO _x , PM _{2.5} , and NMVOC should be reduced by at least 51%, 64%, 53%, and 36%, respectively, by 2030 from the 2012 levels, in order to meet the ambient PM _{2.5} standard.



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Figure 1 Annual average $PM_{2.5}$ mass concentrations in China (a) in 2013, (b) in 2014, (c) in 2015.
(d) Difference of $PM_{2.5}$ concentration between 2015 and 2013.



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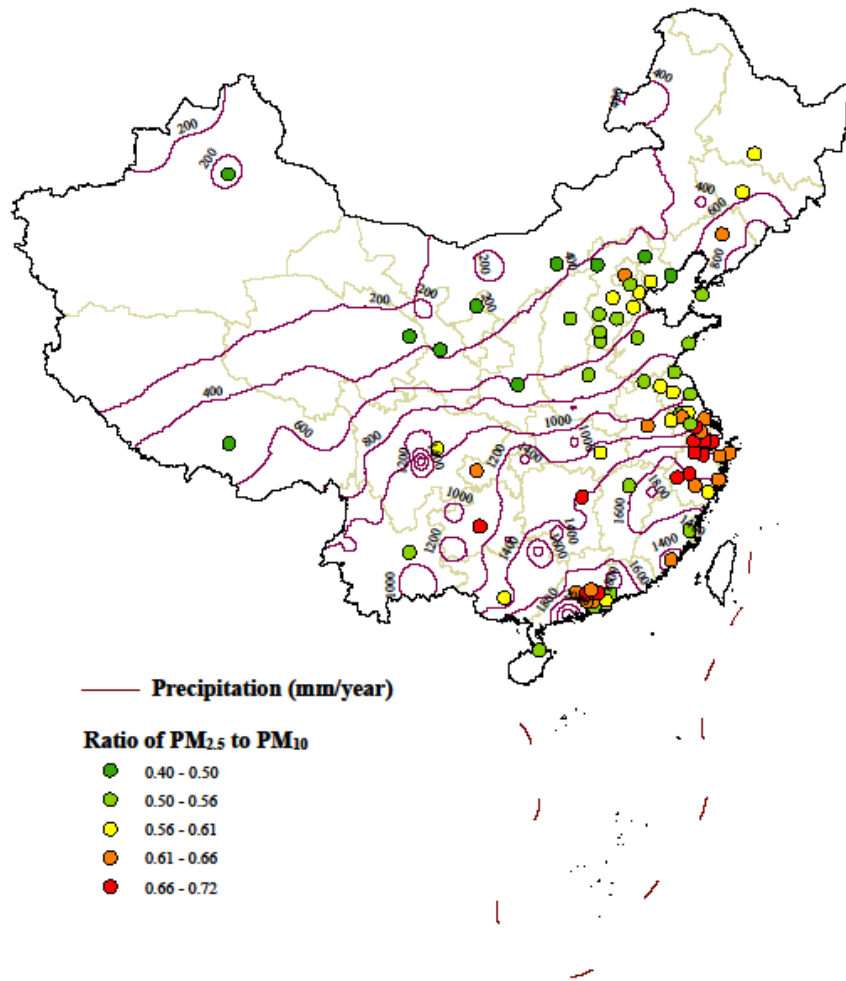
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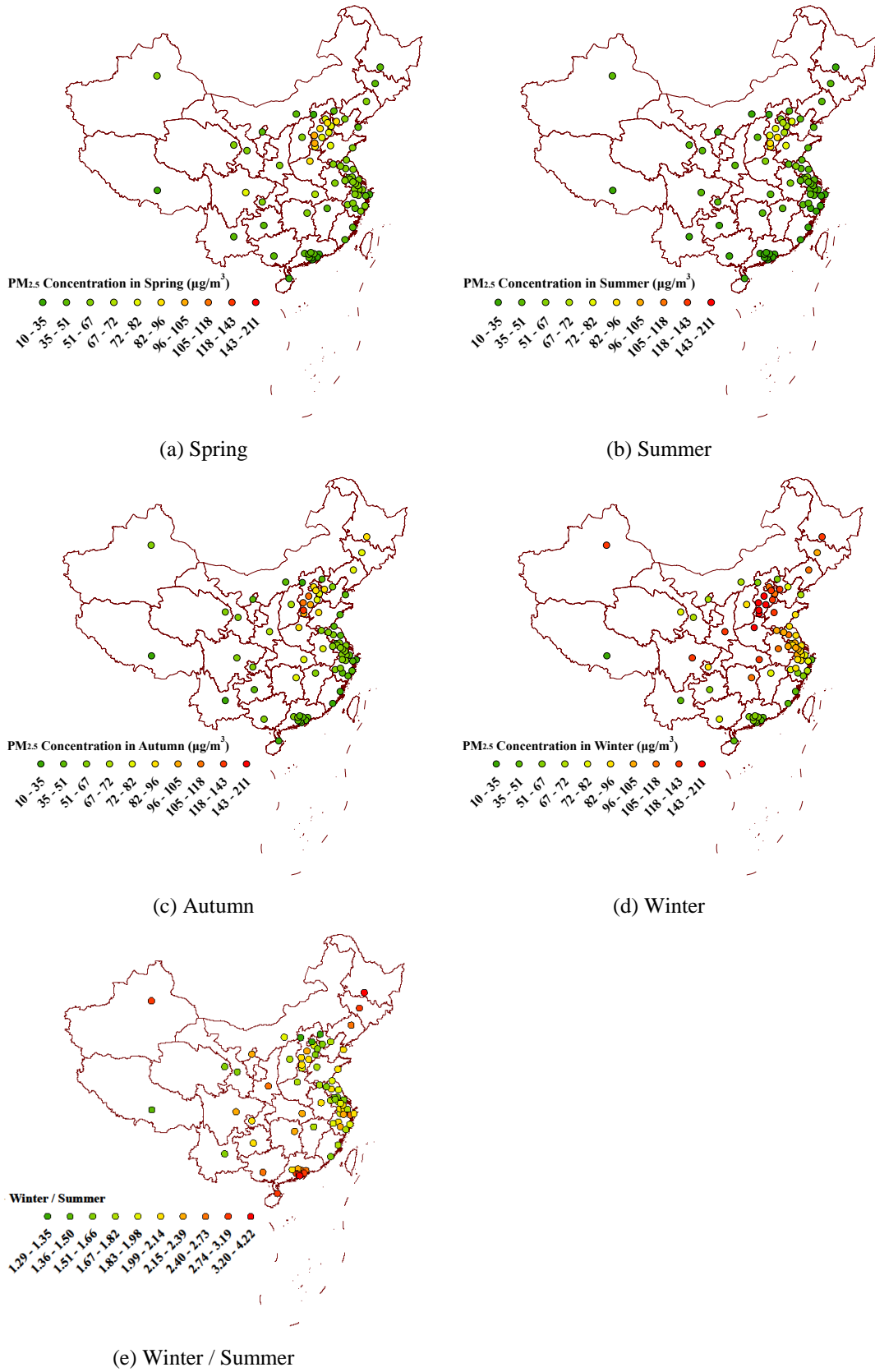
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Figure 2 Annual average PM_{2.5} concentration in Europe and China. The grey line shows the average value of PM_{2.5} concentration of in large and middle-size cities. The whisker and box show the min (max) value and 25 (75)% percentiles, respectively. The red, orange, and green dashed line shows the air quality standard of China and Europe, and the WHO guideline, respectively.



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Figure 3 Ratio of PM_{2.5} to PM₁₀ in China in 2015 and isohyetal line.

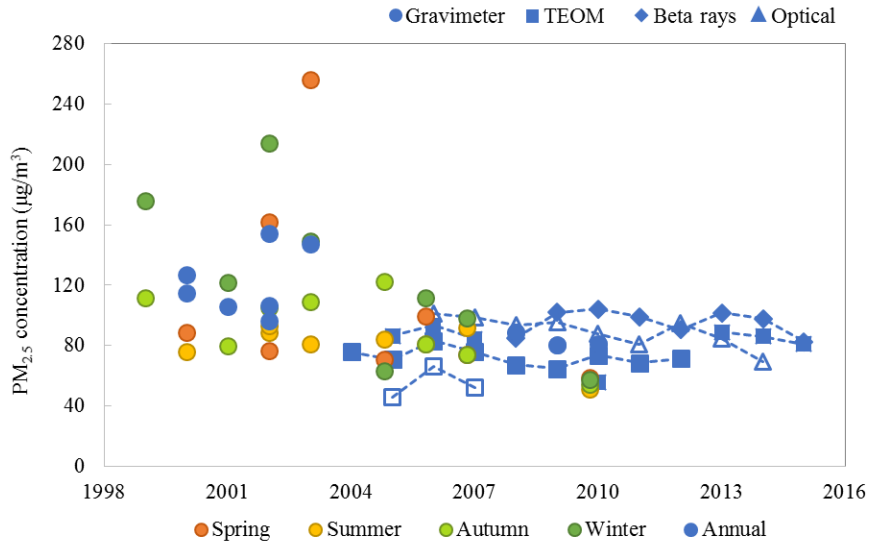


(e) Winter / Summer

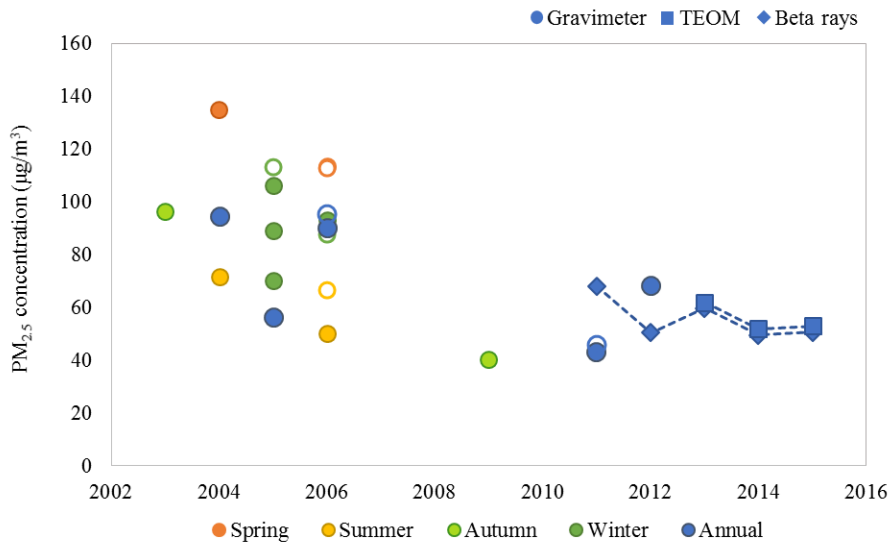
Figure 4 seasonal variation of PM_{2.5} mass concentration

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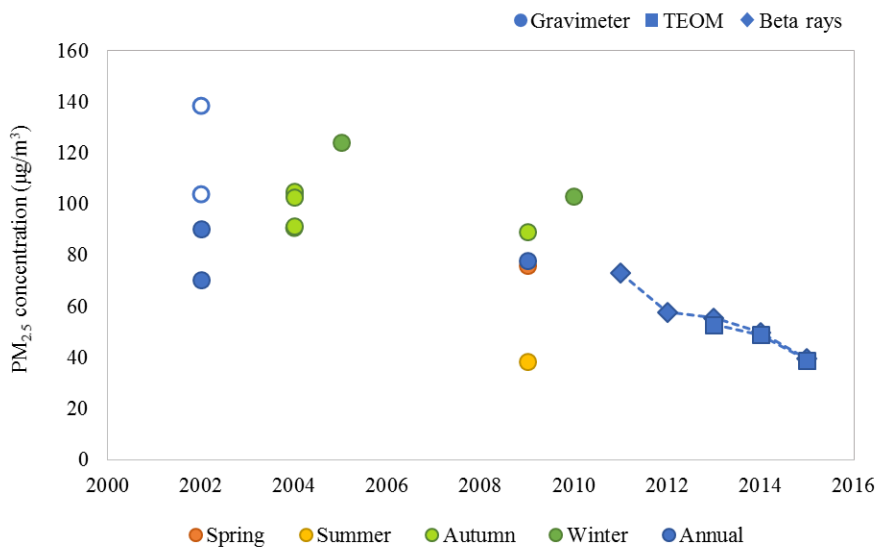
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(a) Beijing

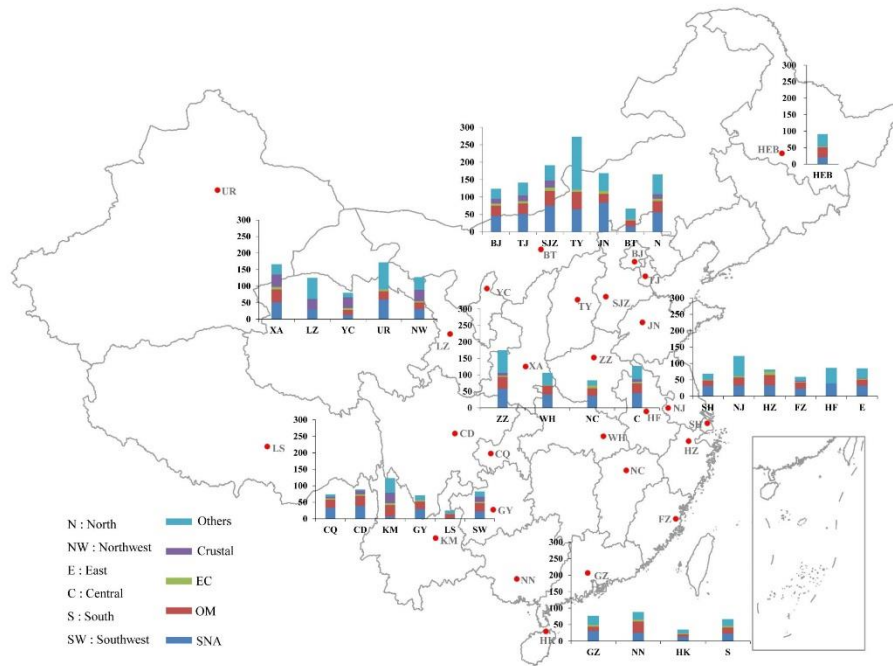


(b) Shanghai



(c) Guangzhou

1238 Figure 5 Interannual trend of average PM_{2.5} mass concentrations in megacities. Symbols with dashed
1239 line shows multi-year studies. Blue represents annual average PM_{2.5} concentration. Orange, yellow,
1240 light green, and dark green represent average concentration of spring, summer, autumn, and winter,
1241 respectively. Circle, square, diamond, and triangle represent measurement methods, i.e., gravimeter,
1242 TEOM, beta rays, and optical method, respectively. Solid and hollow represent urban area and rural
1243 area, respectively.
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Figure 6 Annual average concentrations ($\mu\text{g m}^{-3}$) of PM_{2.5} and its major chemical components in 27 major cities (23 provincial capitals plus 4 municipalities) and different regions over China

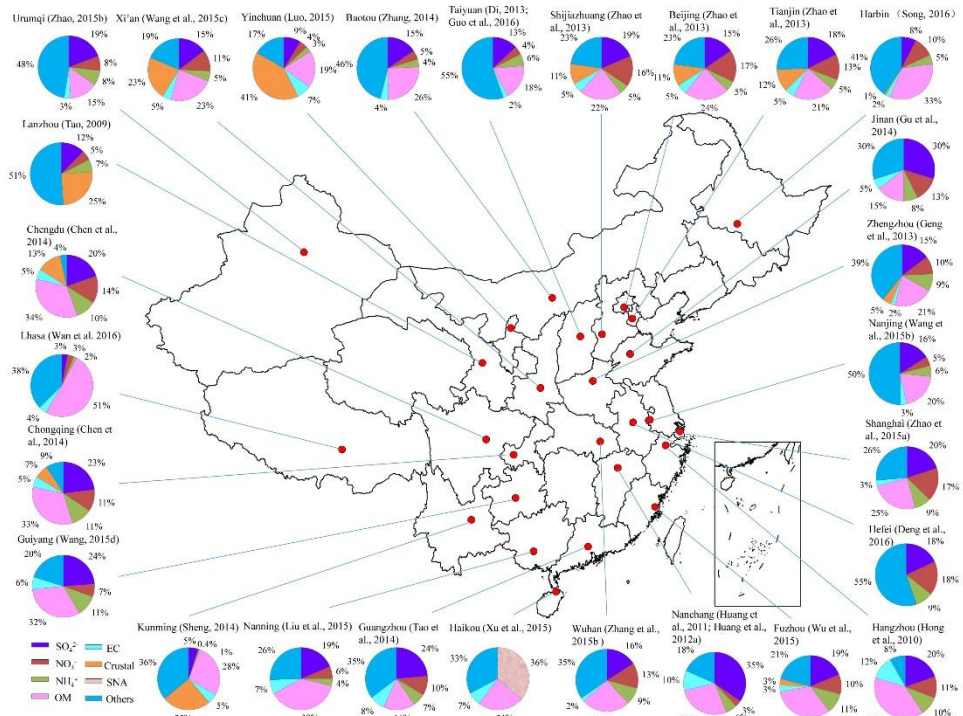
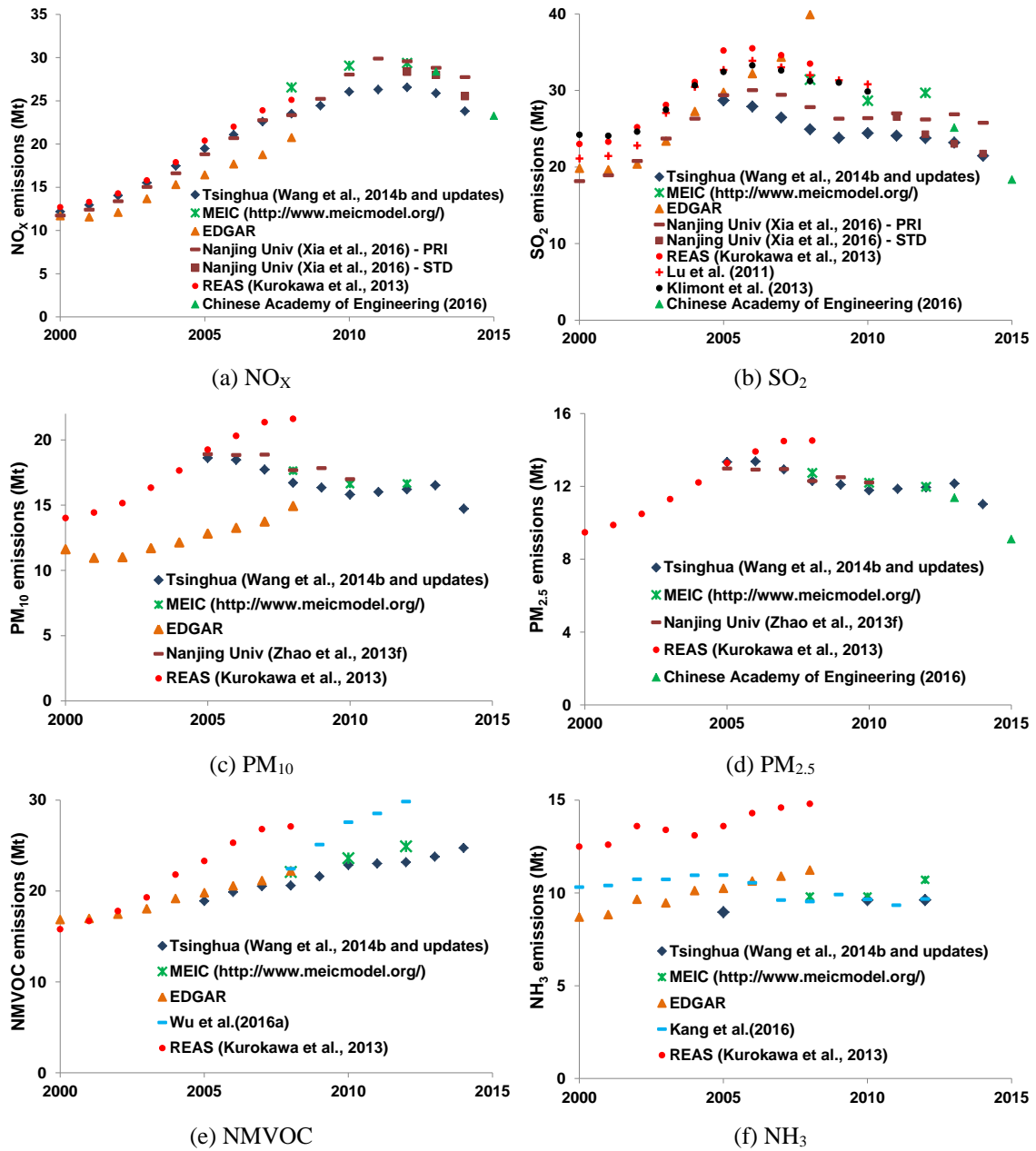
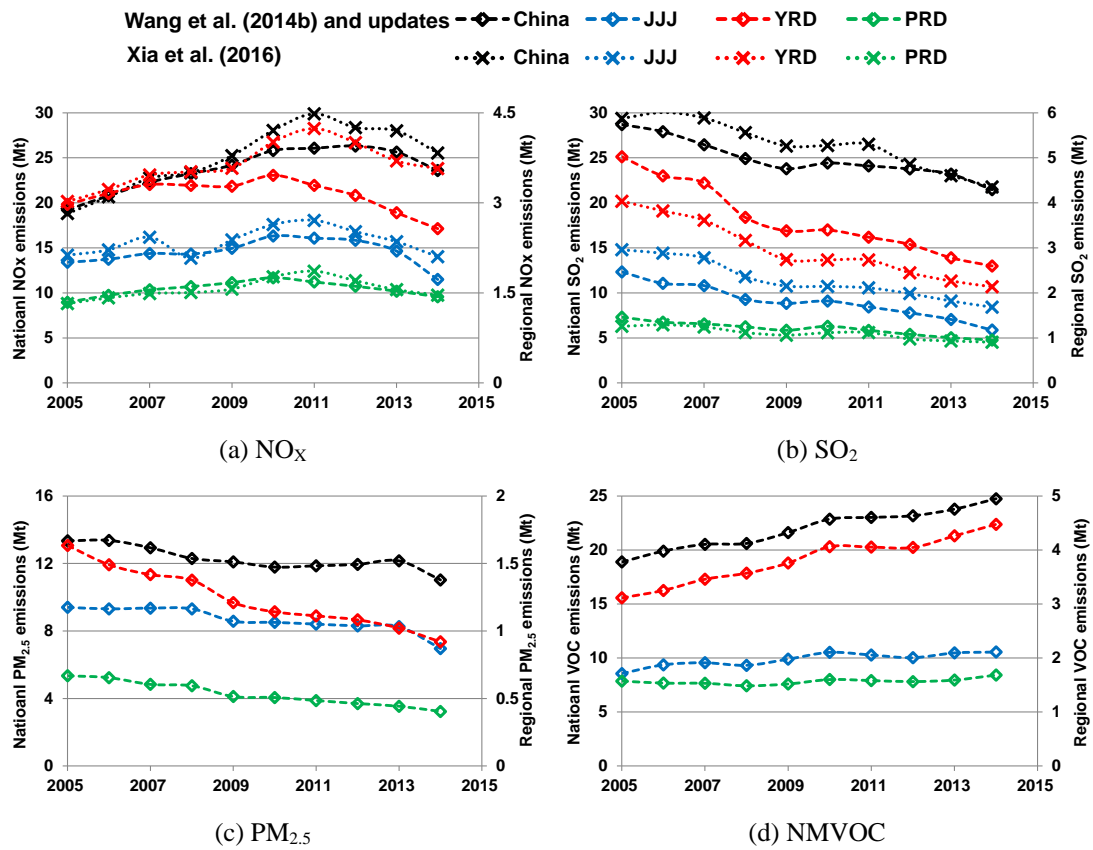


Figure 7 Averaged PM_{2.5} speciation across China

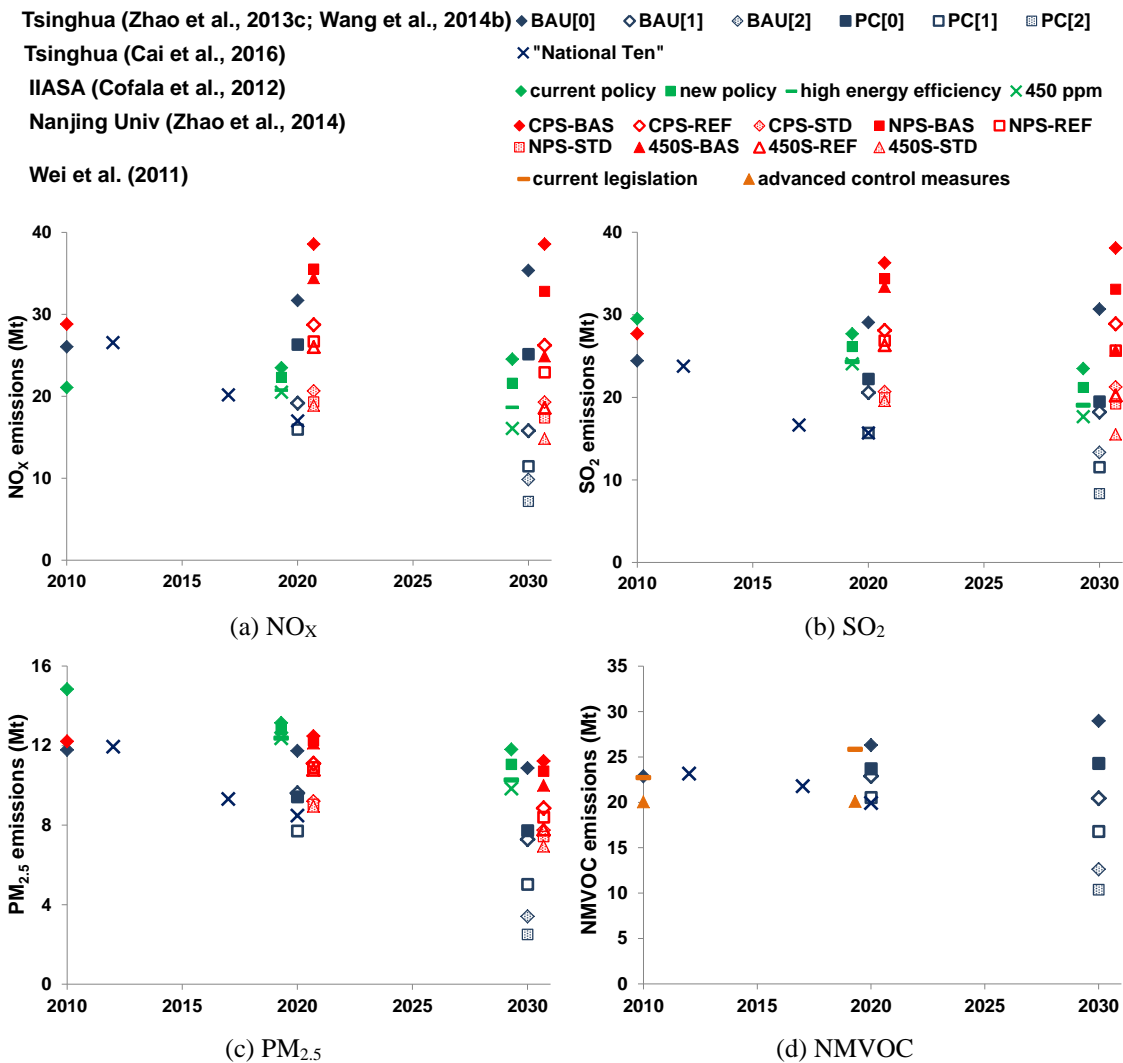
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1252 Figure 8 Emissions of major air pollutants in China during 2000-2015: (a) NO_x , (b) SO_2 , (c) PM_{10} , (d)
 1253 $\text{PM}_{2.5}$, (e) NMVOC, (f) NH_3
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1256 Figure 9 Emissions of major air pollutants in key metropolitan regions of China during 2005-2015: (a)
 1257 NO_x, (b) SO₂, (c) PM_{2.5}, (d) NMVOC. The results are derived from Wang et al. (2014b) and subsequent
 1258 updates, and the STD case of Xia et al. (2016).
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1260 Figure 10 Projections of future emissions of major air pollutants in China up to 2030: (a) NO_x, (b) SO₂,
 1261 (c) PM_{2.5}, (d) NMVOC. The scenarios developed by the same institute are shown with symbols of the
 1262 same color, and since their historical emissions duplicate each other, we show just the historical values
 1263 of one scenario. Some points for the years 2020 and 2030 are shifted a little left or right, in order to
 1264 avoid overlapping representation.
 1265