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Evaluating the real changes of air quality due to clean air actions using a machine learning technique: results from 12 Chinese mega-cities during 2013-2020

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21

22 Abstract

23 China has implemented two national clean air actions in 2013-2017 and 2018-2020, respectively, with the aim of reducing primary emissions and hence improving air 24 quality at a national level. It is important to examine the effectiveness of such emission 25 reductions and assess the resulting changes in air quality. However, such evaluation is 26 difficult as meteorological factors can amplify, or obscure the changes of air pollutants, 27 in addition to the emission reduction. In this study, we applied the random forest 28 machine learning technique to decouple meteorological influences from emissions 29 changes, and examined the deweathered trends of air pollutants in 12 Chinese mega-30 31 cities during 2013-2020. The observed concentrations of all criteria pollutants except 32 O₃ showed significant declines from 2013 to 2020, with PM_{2.5} annual decline rates of 6-9% in most cities. In contrast, O₃ concentrations increased with annual growth rates 33 of 1-9%. Compared with the observed results, all the pollutants showed smoothed but 34 similar variation in trend and annual rate-of-change after weather normalization. The 35

response of O₃ to NO₂ concentrations indicated significant regional differences in 36 photochemical regimes, and the differences between observed and deweathered results 37 provided implications for volatile organic compound emission reductions in O₃ 38 pollution mitigation. We further evaluated the effectiveness of first and second clean air 39 40 actions by removing the meteorological influence. We found that the meteorology can make negative or positive contribution in reducing pollutant concentrations from 41 emission reduction, depending on type of pollutants, locations, and time period. Among 42 the 12 mega-cities, only Beijing showed a positive meteorological contribution in 43 amplifying reductions in main pollutants except O₃ during both clean air action periods. 44 Considering the large and variable impact of meteorological effects in changing air 45 quality, we suggest that similar deweathered analysis is needed as a routine policy 46 47 evaluation tool on a regional basis.

48



50 meteorological influence



53



52 Graphical Abstract

54 **1. Introduction**

55 Air pollution is an urgent problem globally due to its adverse impacts on the environment, human health and climate (Fan et al., 2020; Hadley et al., 2018). It has been well recognized SO₂. 56 NO_2 , CO_3 , $PM_{2.5}$ and PM_{10} are defined as the six criteria pollutants in quantifying air pollution 57 levels (Hu et al., 2015). World Health Organization (WHO) data shows that 9 out of 10 people 58 59 breathe air that contains high levels of these criteria pollutants and which exceeds WHO guideline 60 limits, and it is estimated that 7 million people premature deaths are caused by air pollution worldwide every year (World Health Organization, 2021). In addition, air pollution can reduce 61 62 visibility and affect solar radiation balance directly and indirectly (Li. et al., 2017; Xia et al., 2016), and even give rise to more extreme weather events (i.e. flooding and drought (Cui et al., 2017;
Herrera-Estrada et al., 2018; Tie et al., 2016).

65 In the past few decades, China has experienced rapid industrialization and urbanization. Along with the rapid development of the economy, air pollution has produced a substantial influence on 66 each sector of Chinese society for a long time. In some major areas of China, concentrations of air 67 68 pollutants greatly exceed air quality guidelines for the protection of health recommended by the 69 WHO (Zhang et al., 2015). In China, ambient air pollution has become the fourth largest threat to 70 Chinese health, after heart disease, dietary risks and smoking (Chen et al., 2013; Zhang et al., 2015), 1,565,000 71 and was responsible for to 2,168,000 premature deaths in 2019 72 (http://ghdx.healthdata.org/gbd-results-tool). As a result, air pollution has become a major concern for the public and policy makers (Feng et al., 2017; Guo et al., 2020; Kuerban et al., 2020; Liu and 73 74 Wang, 2020; Ma et al., 2019; Song et al., 2017; Wang et al., 2019a; Wang et al., 2019b; Zhan et al., 75 2018; Zhang et al., 2018). To solve the increasingly serious air pollution problem, China established 76 a national air quality monitoring network that covers major cities since early 2013. The monitoring 77 data include observations of PM2.5, PM10, NO2, SO2, O3 and CO (Wang et al., 2020b). Meanwhile 78 in 2013, the Chinese government implemented the "Air Pollution Prevention and Control Action 79 Plan" (2013-2017), also known as the first Clean Air Action. In 2018, the Chinese government 80 continued to implement the "Blue Sky Protection Campaign" (2018-2020), also known as the 81 second Clean Air Action (Wang et al., 2019c; Xu et al., 2021).

82 At present, these actions have been implemented, and it is of great significance to accurately 83 estimate whether the intervention is working to meet the set targets. However, such evaluation is 84 somewhat difficult as many meteorological conditions can obscure the impact of emission changes 85 on air quality (Vu et al., 2019). Apart from interventions or management efforts to control air 86 pollution, meteorological conditions can also directly or indirectly affect the emission, transport, 87 chemical formation and deposition of air pollutants, thus affecting their concentration in ambient 88 air (Zhang et al., 2015). Variation of meteorological factors can hinder the correct analysis of trends 89 in different air pollutants and may lead to erroneous conclusions about the effectiveness of 90 intervention or management strategies (Grange and Carslaw, 2019). Hence, it is essential to 91 decouple meteorological impacts from trends in ambient air quality data and extract the real changes 92 in air quality driven by policy interventions.

93 Daskalakis et al. (2016) employed chemical transport models to evaluate the response of air 94 quality to emission control measures, which was based on assimilated meteorology to account for 95 the year-to-year climate variability. However, such assessment results will be affected by significant 96 uncertainties in the emission inventory and chemical transport model itself (Gao et al., 2018). 97 Statistical analysis is another commonly used method to decouple the meteorological effects on air 98 quality. Many mathematical analysis approaches or models were developed, which were mainly 99 through data regression to eliminate the impact of varying meteorological variables. Venter et al. 100 (2020) adopted a regression model to evaluate the effects of COVID-19 lockdown on air pollution 101 levels. He et al. (2020) applied a "difference-in-difference" approach to evaluate the impacts of 102 COVID-19 lockdown measures in terms of the Air Quality Index (AQI) and the concentrations of 103 particulate matter. Henneman et al. (2015) employed multiple Kolmogorov-Zurbenko filters and a 104 multi-linear regression model to estimate the effectiveness of air pollution regulations and control 105 measures. Among these models, machine learning models (i.e., the boosted regression trees and 106 random forest algorithms) usually show a better performance than traditional statistical and air

quality models by reducing variance/bias and error in high dimensional data sets, though they fail to interpret the physical mechanism behind the results (Zhang et al., 2020). Particularly, RF has the advantage of not being a "black-box" method where the learning process can be explained, investigated, and interpreted. Recently, Grange et al. (2018) developed a machine learning technique based upon the random forest algorithm to identify the key mitigation measures contributing to the reduction of air pollutant concentrations in Beijing. This technique has been adopted widely and

validated in various environments (Shi et al., 2021; Vu et al., 2019; Zhang et al., 2020).

In this study, we applied the RF machine learning technique proposed by Vu et al. (2019), and systematically studied the trends and characteristics of air pollution in 12 mega-cities in China from 2013 to 2020 based on the latest observational data. The effects of meteorological factors were eliminated from the trends in pollutant levels, and the real performance of the two national clean air actions were comprehensively assessed. The results may be conducive to formulating air quality control policies in China and other developing countries.

120 **2. Method**

121 2.1 Study region and data sources

In this study, we investigated 12 megacities in China, these cities are selected as they are representative mega-cities of China according to their GDP, population and area, and geographical distribution. Also, the air quality data availability is another important reason, as all these 12 megacities started to monitor $PM_{2.5}$ since January 2013. Their geographical locations are shown in Fig. S1, and the summarized information describing each city (i.e. economy, population, area, specific location, etc.) can be found in the Supporting Information Table S1.

128 Since January 2013, the Ministry of Environmental Protection of China (MEPC) has released 129 the real-time air pollution monitoring information for 74 major cities, including for the 6 main 130 criteria pollutants of PM_{2.5}, PM₁₀, NO₂, SO₂, O₃ and CO. In this study, we used air quality 131 observation data and meteorological data over the aforementioned 12 cities from 18th January 2013 132 to 31st December 2020. Table S2 summarized the name and location of monitoring stations in each city from the Chinese national monitoring network, and those sites are mostly distributed in urban 133 134 areas of each city. By averaging the measurements from the monitoring sites within each city, we obtain city-specific dataset that can represent the air quality of each city. Meteorological data, 135 including air pressure (PRS, hPa), temperature (TMP, °C), wind direction (WD, °), wind speed (WS, 136 137 m/s), and humidity (RH, %), were also collected from meteorological stations of each city over the 138 period of 18th January 2013 to 31st December 2020. The detailed information of data source was 139 illustrated in Supporting Information Text S1.

140

141 2.2 Deweathering using the random forest (RF) model

The deweathering technique was first proposed by Grange et al. (2018) to predict the concentrations of air pollutants at a specific measured time point but removing the influence of varying meteorological conditions, which was regarded as "deweathered concentration". Such technique was based on RF regression model. Regression model is a mathematical model for quantitative description of statistical relationship, and it can indicate the strength of the influence of multiple independent variables on one dependent variable. The RF regression model is composed

of hundreds of independent decision tree models and a combination of randomly chosen explanatory 148 149 factors, and the term "Random Forests" is derived from "random decision forests". The random 150 selection process involves 1) variables and data input, and 2) generation of a certain number of decision trees. In the subsequent calculation, each decision tree can provide one intermediate for the 151 152 input variables, and then the intermediate of these decision trees were summarized as the RF 153 regression prediction result. It can be employed to describe the relationship between the 154 concentration of air pollutants by temporal variables (yearly, day of year, day of week, and hourly) 155 and meteorological variables (relative humidity, temperature, atmospheric pressure, wind speed and wind direction). The detailed information of random forest model can be found in Supporting 156 157 Information Text S2 (Wang et al., 2020a).

158 Firstly, we constructed a reliable RF model. The original datasets for the RF model of each city 159 contain the concentration of PM_{2.5}, PM₁₀, NO₂, SO₂, O₃ and CO as well as their predictor variables, 160 including time variables represented by Unix Epoch time, hour (0-23), day of week (Monday to 161 Sunday), and meteorological parameters (wind speed, wind direction, pressure, temperature, and 162 RH). 70% of the original datasets were randomly selected as a training dataset to construct the RF model, using R "normalweather" packages by Grange et al. (2018), and the remaining 30% of the 163 164 original data was employed as testing dataset to validate the performance of the constructed model. 165 The performance of the RF model has been evaluated based on several typical statistical metrics 166 (Emery et al., 2017; Vu et al., 2019), and details of formulas, verification plots and summarized 167 statistical metrics can be found in the Supporting Information Text S3, Fig. S3 and Table S3. These results demonstrated the reliability of the trained model. For example, the coefficient of 168 169 determination r^2 are all above 0.8, and the coefficient of efficiency COE are between 0.6 and 0.9. 170 These validation results from different statistical metrics are overall consistent with previous study 171 (Vu et al., 2019), hence the trained model can be used for subsequent deweathering analysis.

Secondly, we used the validated RF model to eliminate the effects of meteorological factors. 172 Both time variables (month, week, and hour) and meteorological parameters were resampled 173 174 randomly to represent the mean meteorological conditions of a city. Specifically, we resampled 175 meteorological data set from the meteorological conditions of that city for 2013-2020. The input 176 meteorological variables at a particular time point on a particular day can be randomly replaced with 177 data at the same time within a four-week period (i.e., 2 weeks before and 2 weeks after the selected 178 date), which is similar to Zhang et al. (2020). Then the resampled meteorological variables and time 179 variables were fed into the RF model to predict the concentrations of air pollutants. A diagram that 180 describing the above methodology is shown in Fig. S2. This resample and calculation process was 181 repeated 1000 times, and the 1000 predicted concentrations of each air pollutant at specific time 182 point were obtained and then averaged as the final weather-normalized concentration. In this way, 183 the impact of weather variations on air pollutants can be normalized while their seasonal and diurnal 184 variations are retained, which allows us to further study the temporal variations in deweathered 185 concentrations.

186 **3. Results and discussion**

187 **3.1 Observed and deweathered air quality trends during 2013-2020**

Combining the observed and deweathered pollution concentration data, we obtained the annual changing rate (normalized to the year of 2013) of the six criteria pollutants in the 12 mega-cities from 2013 to 2020 by a linear regression, which are shown in Fig. 1. Detailed trends about the monthly and yearly averaged values for both observed and deweathered concentrations can be found in Supporting Information Fig. S4-S5.

193 As shown in Fig. 1, the overall concentrations of PM_{2.5} observed in 12 mega-cities maintained 194 a general downward trend, while several cities experienced an uptick in specific years, such as 195 Shijiazhuang in 2016 and Shanghai in 2015. Specifically, the annual decline rate of PM_{2.5} observed 196 across the 12 mega-cities has been around 6~9% per year, and the decline rates in Beijing, 197 Shijiazhuang, Nanjing and Hangzhou are larger than for the other cities. The trends in the weather 198 normalized concentrations and annual decline rates of PM2.5 in most of the mega-cities are very 199 similar to that of observed trends and annual decline rate, with the exceptions of Beijing and 200 Lanzhou with 2% lower deweathered annual decline rate compared to that of observed PM_{2.5}.

Apart from Shijiazhuang, Lanzhou and Chengdu, the concentration of observed PM₁₀ in many cities increased in 2014 and then declined subsequently. The concentrations of observed PM₁₀ in Shijiazhuang, Lanzhou and Chengdu increased slightly in 2016 and maintained a downward trend in the other years. The annual decline rates of observed PM₁₀ are around 6%~8% in Beijing, Tianjin, Shijiazhuang, Nanjing, Wuhan, Chengdu and Chongqing, and 4%~6% in Shanghai, Hangzhou, Guangzhou, Shenzhen and Lanzhou. After weather normalization, the deweathered PM₁₀ showed overall similar annual decline rates to the observed, consistent with the case of PM_{2.5}.

208 Large variability in the trends for observed NO₂ concentrations were found among the 12 cities. 209 Specifically, Chongqing and Lanzhou maintained an upward trend from 2013 to 2017, and began to decline from 2018. By contrast, other cities showed a downward trend of observed NO₂ since 2013 210 211 or 2014 with some fluctuations in specific years. In 2014, Tianjin, Shanghai, Lanzhou and 212 Chongqing experienced a significant NO₂ uptick, while Nanjing, Hangzhou, Chengdu and Beijing 213 experienced a sightly increase. In 2017, Tianjin, Guangzhou and Wuhan experienced a significant 214 uptick while Nanjing and Shanghai experienced a sightly increase. Overall, the annual rate of decline of the observed NO₂ is highest in Beijing (7% per year), followed by Chengdu, Shenzhen 215 216 and Wuhan (around $4\% \sim 6\%$), and the rates of decline for other cities are all below 4%. 217 Comparatively, large fluctuations of the observed NO_2 in specific years were smoothed after the weather normalization. For example, the NO₂ increase phenomenon (i.e. Hangzhou and Chengdu 218 219 in 2014; Tianjin and Guangzhou in 2017) was not observed in deweathered trends. The annual 220 decline rates of NO₂ after the weather normalization were overall similar to that of the raw observed concentrations, except for Shanghai showing a clear difference between observed and deweathered 221 222 (-0.4% vs. -2.1%).

The concentration of observed O₃ in most of the mega-cities showed an upward trend until 224 2017, except for Tianjin and Shijiazhuang. Specifically, Lanzhou, Shijiazhuang and Tianjin showed 225 the highest annual rate of increase in observed O₃ at around 8%, followed by Nanjing (6%), while 226 other cities showed lower rates of increase (below 4%). Similar to NO₂, the observed O₃ fluctuations in specific years were smoothed after the weather normalization. Overall, both observed and deweathered O₃ concentrations have increased for all the cities, with annual increase rates of 1-9%.

229 The reduction in observed SO₂ concentrations was the most significant among the six criteria pollutants. From 2013 to 2020, all 12 mega-cities maintained a large SO₂ reduction rate in the first 230 231 few years, which then gradually reduced. Particularly for Wuhan, Chongqing, Shenzhen, Lanzhou 232 and Hangzhou, the observed SO₂ concentrations tended to flatten out since 2016. Overall, the annual rates of decline of observed SO₂ in most of the mega-cities exceeded 10%, with Beijing, Tianjin and 233 234 Shijiazhuang showing the largest rate of decline (around 12%), while the rate of decline for 235 Shenzhen is the lowest (6%). Compared to the observed results, almost all the mega-cities showed 236 a similar trend and rate of decline in deweathered SO₂ results, except Lanzhou with a smaller 237 decrease of deweathered trend than that observed since 2019.

Generally, the concentration of observed CO in all mega-cities showed a trend of declining 238 239 concentrations, with some fluctuations for specific years. For example, Lanzhou and Chongqing 240 showed a brief increase in observed CO in 2014, while Nanjing and Shijiazhuang also showed a 241 significant increase of CO in 2016. Overall, Beijing, Tianjin, Shijiazhuang, Shenzhen and Chengdu showed a relatively large CO annual reduction rate of 6%~8%, while the reduction rate for other 242 243 cities was around 2%~4%. Similar to other pollutants, the fluctuations of CO in specific years were 244 smoothed after the weather normalization (i.e., Lanzhou and Chongqing in 2014; Nanjing in 2016). 245 The annual rate of decline of CO after the weather normalization is very similar to that obtained 246 from the observed values, with differences generally within 1%.

247 We further define the index ω to quantify the extent to which concentrations of the six criteria 248 pollutants in different regions are affected by meteorological conditions. As meteorology may have 249 positive or negative influences in different seasons (Vu et al., 2019), we used absolute difference 250 between observed and deweathered values to prevent an offsetting effect arising between different 251 seasons. The absolute difference values from 96 months between 2013 and 2020 are averaged, as 252 shown in Equation (1).

253
$$\omega = \frac{\sum_{i=1}^{96} \frac{\left|C_{i,observation} - C_{i,model}\right|}{C_{i,observation}}}{96} \tag{1}$$

254 The ω values for the six criteria pollutants across 12 mega-cities are shown in Fig. S6 and the overall results are discussed here. $PM_{2.5}$, PM_{10} and O_3 were most affected by meteorological 255 conditions (ω between 10%~15%), followed by NO₂ and SO₂ (ω ~10%). Compared with other 256 257 primary gaseous pollutants, it seems that $PM_{2.5}$ and PM_{10} are more sensitive to meteorological 258 influences. The likely reasons are 1) long-range transport is important source of aerosol, which is 259 mainly controlled by the wind field; 2) temperature and humidity are also important meteorological factors that can affect secondary aerosol formation and chemical composition. CO shows the lowest 260 ω value of around 5%, which may be explained by its long atmospheric lifetime that has least 261 influence from meteorological conditions. Among 12 mega-cities, the air pollutants (i.e., PM_{2.5}, 262 263 PM_{10} , SO₂ and CO) in Beijing are most affected by meteorological conditions, which is likely partly 264 due to the unique topography in Beijing as the Yanshan Mountains and Taihang Mountains surround 265 the city to the north and west. For example, northern winds tend to bring clean air to Beijing while 266 southerly winds do the opposite (Liang et al., 2017; Zhang et al., 2015). Here we further performed the 72 hours backward trajectory analysis at Beijing from 2013 to 2020 by employing the hybrid 267

single particle Lagrangian integrated trajectory model (Hysplit) with the global data assimilation system (GDAS). The trajectory clustering analysis showed that more than 50% long-range airmass arriving in Beijing were mainly originated from the northwest direction and passing through Yanshan Mountains and Taihang Mountains (Fig. S7). As mentioned previously, our RF model results suggested that Beijing is most affected by meteorological conditions among the 12 megacities, which may be explained by both unique topography in Beijing and frequent long-range airmass from northwest direction. By contrast, there were no significant city-to-city differences in

275 the relative sensitivity to meteorological conditions for NO_2 and O_3 .

276 **3.2 Response of O₃ to NO₂ concentrations**

277 It is well established that both NOx and volatile organic compounds (VOCs) are the precursors of O_3 , and understanding the O_3 -precursor relationship is important for developing effective O_3 278 control strategies (Blanchard, 2000; Hidy, 2000; Li et al., 2019; Lu et al., 2018). Here we explore 279 280 the O_3 -NOx relationship for each city by analysis of the monthly averaged O_3 and NO_2 281 concentrations (as volume mixing ratios, i.e. converted to ppbv to retain the NO₂-O₃ photostationary 282 steady state stoichiometry) across all 96 months during 2013-2020 (Fig. 2). Data for most of the 283 cities (except Guangzhou and Shenzhen) showed clear negative correlation between O_3 and NO_2 , 284 suggesting that NOx reduction can lead to the increase of O₃. According to the well-known titration reaction (NO + O₃ \rightarrow NO₂ + O₂), neglecting other processes an increase of 1 ppbv NO₂ would lead 285 286 to 1 ppbv consumption of O_3 , corresponding to a slope of -1 in the O_3 vs. NO_2 space. However, 287 this slope value may vary due to the complex non-linearity of the O₃-VOC-NOx chemistry, which 288 provides an indicator of the O₃ formation chemistry (i.e., O₃ formation is more or less sensitive to 289 NOx reduction).

290 As shown in Fig. 2, the regression slope indicates how O_3 responses to the changes of NOx 291 concentration for each city. Interestingly, it can be seen from observed category that the slope values 292 showed significant regional differences with variations between cities. For example, the slope 293 values for BTH region (Beijing-Tianjin-Hebei, Shijiazhuang is the capital of Hebei province) ranged 294 from -1.42 to -1.36, while YRD region (Yangtze River Delta, including Shanghai, Hangzhou and 295 Nanjing) ranged from -1.09 to -0.08. By contrast, data from the PRD region (Pearl River Delta, 296 including Guangzhou and Shenzhen) did not show any clear negative relationship between O₃ and 297 NO_2 (with slope of ~0). The above information is indicative of regional differences in O_3 -precursor 298 relationship. Here we further assume that the slope value of -1 as the threshold, with values 299 significantly above or less than -1 as more or less sensitive to NOx reduction respectively. In 300 addition, we compare our fitted slope values in O₃-NO₂ space with the O₃ formation regime 301 identified from HCHO/NO₂ ratios in Li et al. (2021) for the three key regions. As shown in Table 302 S4, these results are overall consistent, which further demonstrates that O₃ formation in PRD is more 303 sensitive to NOx reduction, while O_3 formation in other regions is less sensitive to NOx reduction. 304 Note that the above O₃-precursor relationship derived from the fitted slope value in the O₃ vs. NO₂ 305 space is qualitative as a relative concept, and further detailed evaluation using a quantitative method 306 (e.g. chemical transport model) is still needed.

307 Compared with the observed concentrations, the overall range in deweathered concentrations 308 for O_3 and NO_2 are narrowed, suggesting that extreme conditions due to meteorology have been 309 removed. Note that almost all the cities showed slightly decreased slopes (O_3 vs NOx) with improved R^2 after weather normalization, which implies a trend of O₃-precursor relationship towards less NOx sensitive condition. This suggests that using deweathered data more clearly demonstrates the need for VOC reductions – and the limitations of NOx-reductions alone – in reducing regional O₃ concentrations. The detailed reason for this phenomenon is unclear. However, it implies that the VOC emission reduction should play a more important role for O₃ pollution mitigation in most cities if the meteorological influence is considered.

316 **3.3 Evaluating the effectiveness of two national clean air actions**

317 **3.3.1 First clean air action (2013-2017)**

318 A series of policy, control measures and action plans were proposed for the first clean air action, 319 with a particular focus on Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River Delta 320 regions, attempting to reduce PM_{2.5} by 25%, 20%, and 15%, respectively. Table S5 summarized the 321 main mitigation measures that implemented in China and other key regions during the clean air 322 actions (Zhang et al., 2019; Zheng et al., 2018). Generally, these measures are overall similar among 323 different regions, while there are some region-specific measures according to the differences in 324 energy utilization and industry distribution for each region. According to the recent emission 325 inventory (Zhang et al., 2019), China's anthropogenic emissions have been decreased by 59% for SO₂, 21% for NOx, 23% for CO, 36% for PM₁₀, and 33% for PM_{2.5} during 2013–2017. To evaluate 326 327 the effectiveness of the first clean air action, we summarized the results from the 12 mega-cities as 328 derived from observations before and after weather normalization, as shown in Fig. S8 and Table 1.

329 It is evident that the first clean air action period shows significant reductions in PM_{2.5} concentration, with both observed and deweathered PM2.5 concentrations reducing by 34% on 330 331 average the 12 mega-cities. The reductions indicated the effectiveness of mitigation efforts, such as 332 strengthening industrial emission standards and replacing residential coal with electricity and 333 natural gas (Zheng et al., 2018). The reduction percentages of PM_{2.5} in Beijing and Shijiazhuang 334 reduced greatly after weather normalization, suggesting that meteorological conditions made a 335 positive and significant contribution in reducing $PM_{2.5}$ in Beijing and Shijiazhuang over this time 336 period (see Table 1), while anthropogenic effects (i.e. impacts of emissions) did not reduce as much as the observations suggested (Li et al., 2020). In contrast, meteorological conditions made a 337 338 negative contribution in suppressing reductions in PM_{2.5} in Shanghai, Hangzhou, Wuhan and 339 Chengdu, as the magnitudes of reduction increased after weather normalization, which is consistent 340 with the conclusion of Xiao et al. (2021).

341 Although all the 12 mega-cities showed a downward trend in observed PM_{10} from 2013 to 2017, the magnitudes of the reduction varied widely (0.8% - 45%) among different cities and were 342 343 lower than PM_{2.5} in average (25% vs. 34%). This may partly be attributed to the greater importance 344 of natural primary emission sources to PM_{10} , including dust storms from the desert areas in north 345 and northwest of China (Li et al., 2017). The rates of reduction in deweathered PM_{10} in Tianjin, 346 Guangzhou, Wuhan and Chongqing were larger than the observed, while Beijing and Lanzhou 347 showed the opposite characteristic. This indicates that the meteorological conditions made a 348 negative contribution in suppressing reductions in PM₁₀ in Tianjin, Guangzhou, Wuhan and 349 Chongqing, and a positive contribution in amplifying reductions in Beijing and Lanzhou. In general, the influences on PM_{10} from meteorological conditions are limited compared with anthropogenic emission reduction.

 SO_2 is mainly emitted from the coal-fired source, and the reductions in SO_2 concentration are 352 in line with the coal-fired emission control measures (Li et al., 2020). All the 12 cities showed an 353 354 obvious reduction in observed SO₂ from 2013 to 2017, which suggests that a remarkable SO₂ 355 emission reduction was achieved in the first clean air action period, as all the 12 cities showed an obvious reduction in observed SO_2 from 2013 to 2017, and this is mainly due to the policy measures 356 357 in phasing out of coal-fired boilers (Zheng et al., 2018). The reduction magnitudes of the observed 358 SO₂ ranged from 27% (Shenzhen) to 70% (Beijing), with an average of 56%. All the 12 mega-cities 359 showed very similar values (difference within 4%; see Table 1 and Fig. 3) between deweathered and 360 observed SO₂ reduction magnitudes over 2013-2017, which implies that the meteorological effects 361 can be negligible in obscuring changes in SO₂.

362 Some cities (e.g. Tianjin, Shanghai, Guangzhou, Lanzhou and Chongqing) showed 2.7%~47% 363 increases in observed NO₂ from 2013 to 2017, while other cities showed 8.2%~18% reductions in 364 the same period. The increase in observed NO₂ in some cities is mainly due to the increased vehicle emissions, as indicated by the rapid increase of car ownership in recent years (Song et al., 2018). 365 366 After weather normalization, the increases in NO₂ in some cities (i.e. Tianjin, Shanghai and Lanzhou) 367 were weakened and even reversed to become decreases (i.e. Guangzhou), while the magnitude of NO₂ reduction in Nanjing and Wuhan were enlarged. This suggests that the meteorological 368 369 conditions made a negative contribution in obscuring NO2 reductions in these cities. The effectiveness in reducing deweathered NO₂ was inferior to that of SO₂, which implies that the NOx 370 371 from vehicle emissions were not as effectively controlled, as NOx emissions were from both coal-372 fired power plants and vehicle sources (Meng et al., 2018). Therefore, more effective control 373 measures on vehicle emissions are needed (Lin et al., 2021).

As for O₃, most of the mega-cities (10 out of 12, except Guangzhou and Wuhan) showed an 374 375 increase of observed O₃ from 2013 to 2017 with a range from 14% to 60%. After the weather 376 normalization, the increase magnitudes of O_3 in Beijing, Tianjin and Shijiazhuang were lowered, 377 suggesting that the increase of observed O₃ in these cities was partly due to the negative contribution 378 of meteorological influence. As many previous studies have suggested that VOC-limited O₃ 379 formation regimes dominate the urban areas of China (Ding et al., 2013), the decline in the NOx 380 emission during the first clean air action can be regarded as a significant contributor to increasing 381 O₃ concentration (Lin et al., 2021; Sun et al., 2019). Therefore, we believe that meteorology played 382 an important but not dominant role in increasing the observed O_3 concentrations, consistent with 383 previous studies (Chen et al., 2020a).

384 Most of the mega-cities (8 out of 12) showed 2.6%~31% reductions in observed CO from 2013 385 to 2017. By contrast, Shanghai, Nanjing, Lanzhou and Chongqing showed increases in observed CO from 1.6% to 29%. After weather normalization, the increases in CO were significantly 386 weakened (i.e. Lanzhou) or even reversed (i.e. Shanghai and Nanjing). This suggests that 387 388 meteorological conditions made a negative contribution in obscuring CO reductions in these cities. 389 In general, the effectiveness in reducing CO varied in different cities, demonstrating that policy 390 effectiveness in some cities (i.e. Lanzhou) should be improved. The different trend of CO and NO2 391 may be partly due to the limited implementation of emission reduction measures in Lanzhou, as 392 Lanzhou is an underdeveloping mega-city in the northwest of China with the lowest GDP among 393 the 12 cities (see Table S1). In addition, the unique valley topography in Lanzhou is not conducive

to the diffusion of air pollutants (Chen et al., 2020b), which may frequently offset the emission reduction efforts.

396 3.3.2 Second clean air action (2018-2020)

Continuing the efforts of first clean air action, the second clean air action (2018-2020) aimed to significantly reduce the total emission of major air pollutants, comparing with the base year of 2015 (Li et al., 2020). The concentrations and changes between 2015 to 2020 from 12 mega-cities before and after weather normalization are illustrated in Fig. S8. In addition, we also summarized the relative changes (%) of six criteria pollutants during 2015-2020 and 2018-2020 in Table 1.

402 From 2015 to 2020, 12 mega-cities have achieved significant reductions in observed PM_{2.5} 403 concentrations, ranging from 27% (Lanzhou) to 52% (Beijing) with an average of 39%. After the 404 weather normalization, the reductions in deweathered $PM_{2.5}$ concentrations ranged from 20% to 43% 405 (with an average of 36%). The reductions in deweathered PM_{2.5} were significantly lower compared 406 to the reduction in the observed PM_{2.5} in Beijing, Shenzhen and Lanzhou, suggesting that 407 meteorological conditions made a positive and significant contribution in enhancing reductions in 408 PM_{2.5} in these cities during time period of the second clean air action. By contrast, meteorological 409 conditions made a negative contribution in $PM_{2.5}$ reduction at Shijiazhuang, as the magnitude of 410 reduction increased after weather normalization.

411 Compared with the base year of 2015, all the 12 cities showed an obvious downward trend of 412 observed PM_{10} in 2020, and the reduction ranged from 27% (Guangzhou and Lanzhou) to 47% 413 (Beijing). After the weather normalization, the magnitude of reduction ranged from 17% (Lanzhou) 414 to 45% (Wuhan). The downward trends for PM_{10} in Beijing, Shanghai, Nanjing, Shenzhen and 415 Lanzhou were weakened greatly, indicating that meteorological conditions made a positive 416 contribution in the control of PM_{10} in these cities, amplifying the effect of emissions changes.

417 Among the 12 mega-cities, only Lanzhou showed an increase (0.6%) in observed NO₂ from 418 2015 to 2020, while the other 11 cities showed reductions with magnitudes ranging from 4.6% 419 (Tianjin) to 39% (Beijing). After the weather normalization, the increase of NO₂ in Lanzhou was 420 enlarged from 0.6% to 6.8%, indicating the inadequate effort of NOx reduction policies at this 421 location. As shown in Fig. S8 and Table 1, compared to the observed concentrations, the reductions 422 in deweathered NO₂ in Tianjin and Shanghai were enlarged greatly, suggesting a negative 423 contribution of meteorological conditions in offsetting emission-change-driven NOx reductions in 424 these cities.

All the 12 mega-cities showed an obvious decrease in SO_2 from 2015 to 2020, and the reductions of observed SO_2 ranged from 27% to 75%, with 9 cities (except Guangzhou, Shenzhen and Lanzhou) higher than 50%. The SO_2 reduction magnitudes in Beijing, Nanjing and Lanzhou were lowered greatly after the weather normalization, suggesting that the meteorological conditions made a positive contribution in amplifying the reduction of SO_2 in these cities.

430 8 cities (except Shanghai, Hangzhou, Shenzhen and Chengdu) showed an obvious increase of 431 observed O_3 from 2015 to 2020, with the increase ranging from 6.6% to 39%. After weather 432 normalization, only Hangzhou still showed a decrease in O_3 from 2015 to 2020, with magnitude 433 change of only 1.4%. The downtrends in Shijiazhuang and Lanzhou were weakened, which means 434 that meteorological conditions made a negative contribution in the suppressing reduction of O_3 in 435 these cities. The performance of different cities varied widely when compared with the first clean 436 air action. It shows that some cities have made significant improvements in O_3 control during the 437 second clean air action, while O_3 control is still a serious challenge for all these cities and further 438 efforts are needed.

All the 12 cities showed obvious decrease in observed CO from 2015 to 2020, implying the remarkable emission reduction achieved through the second clean air action. The reductions of observed CO ranged from 19% (Hangzhou) to 50% (Beijing), with an average of 29%. By contrast, the reductions of deweathered CO ranged from 16% (Wuhan) to 43% (Beijing), with an average of 27%. The reductions of CO in Beijing, Lanzhou and Wuhan were weakened greatly after weather normalization, suggesting that meteorological conditions made a positive contribution in amplifying effects of the control of CO in these cities.

Here we also compare the overall reduction effectiveness from the average of 12 cities for both clean air actions, with the periods of 2013-2017 and 2018-2020. As shown in Table 1, both periods maintained similar reduction strength for $PM_{2.5}$, PM_{10} and SO_2 . Interestingly, it seems that the second clean air action made greater efforts in reducing NOx and CO, which may explain the slight decrease of observed O₃ during 2018-2020. This implies that the increasing trend severe O₃ pollution can be mitigated through continuous emission reduction.

452 We further summarize the difference between the observed and deweathered relative changes 453 (from values shown in Table 1 and Fig. S8) for each city and six criteria pollutants. A positive value 454 of these difference can indicate the negative contribution made by meteorological factors and vice 455 versa. As shown in Table 1 and Fig. 3, a negative meteorological contribution was observed in most of the cities during the first clean air action, while most of the cities were tended to show a positive 456 meteorological contribution during the second clean air action. However, the difference between the 457 458 observed and deweathered relative changes are mostly below 5%, which suggests that the 459 enhancement or reduction effect of meteorological are overall not significant for each city and six 460 criteria pollutants. This demonstrates that the efforts from emission reduction remain the major 461 driving force for the concentration changes of pollutants. We believe that these positive or negative 462 contributions may be related to many factors, such as the periodic fluctuations in climate and the 463 specific weather characteristics at each city, which still needs further investigation for more detailed 464 explanation. Among the 12 mega-cites, only Beijing showed a positive meteorological contribution 465 in reducing the main pollutants except O₃ during both the first and second clean air actions. This 466 suggests that the improvement of air quality in Beijing was contributed from both emissions 467 reductions of the two clean air actions and favorable meteorological conditions at the same period.

468 **4.Conclusion**

469 In this study, we employed the RF machine learning technique to decouple meteorological factors impacting ambient air quality data for 12 Chinese mega-cities during the period 2013-2020, 470 471 in order to extract the real changes of air quality from large-scale emission reductions in recent years. 472 With the exception of O₃, the observed concentrations of all the criteria pollutants showed 473 significant reduction from 2013 to 2020, with an annual rate of decline for PM_{2.5} in most cities of 474 6-8%, while O₃ showed an annual rate of increase of 1-9%. Compared with the observed results, all 475 the pollutants showed smoothed but similar trends and rates of decline after weather normalization. 476 We further quantify the extent to which these six criteria pollutants that are affected by

meteorological conditions, and the results show that PM_{2.5}, PM₁₀, SO₂ and CO in Beijing are most 477 478 sensitive to meteorology among the 12 mega-cities. By contrast, the responses of NO_2 and O_3 to 479 meteorology did not show significant city-to-city difference. Significant regional differences of 480 ozone formation chemistry were qualitatively determined through the fitted slopes in the O_3 vs. NO_2 481 spaces. Specifically, the O₃ formation in Pearl River Delta (PRD) region is more sensitive to NOx 482 reduction, while O₃ formation in other regions is less sensitive to NOx reduction. The deweathered 483 results emphasize the importance of VOC emission reductions in O₃ pollution mitigation in most 484 cities if the meteorological influence is removed.

485 We further evaluate the effectiveness of first and second clean air actions by removing the 486 meteorological influence. We find that both first and second clean air actions reduced most of the 487 pollutants significantly, excepting the increase of O₃. However, meteorology can play negative or 488 positive role by suppressing or amplifying changes due to emission controls respectively, which 489 ranges from -9% to 16% depending on specific conditions, such as the type of pollutants, locations, 490 and time period. For example, a negative meteorological contribution was observed in most of the 491 cities during the first clean air action, while most of the cities were tended to show a positive meteorological contribution during the second clean air action. Among the 12 mega-cites, only 492 493 Beijing showed a positive meteorological contribution in reducing the main pollutants except O_3 494 during both the first and second clean air actions. Considering the significant scope for 495 meteorological effects to change air quality, and obscure the effectiveness of policy actions, we 496 suggest that such deweathered analyses are routinely undertaken on a regional basis.

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499 **Conflicts of interest**

500 The authors declare that they have no conflicts of interest.

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504 CRediT authorship contribution statement

Yong Guo: Formal analysis, Investigation, Software, Visualization, Writing- Original draft
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Table 1. The relative changes (%) of six criteria pollutants from 2013 to 2017, 2015 to 2020, and

661 2018 to 2020 in 12 mega-cities.

	Observed concentration							Deweathered concentration					
		PM _{2.5}	PM10	NO ₂	SO_2	O ₃	CO	PM _{2.5}	PM10	NO ₂	SO_2	O3	CO
	2013-2017	-33	-20	-17	-70	14	-31	-27	-15	-10	-66	1	-24
Beijing	2015-2020	-52	-47	-39	-71	6.6	-50	-41	-39	-36	-65	7.2	-43
	2018-2020	-23	-30	-26	-35	1.3	-22	-19	-28	-25	-25	3.2	-21
	2013-2017	-27	-8.9	23	-60	37	-2.6	-26	-12	17	-52	24	-8
Tianjin	2015-2020	-31	-42	-4.6	-70	39	-34	-31	-42	-15	-68	38	-34
	2018-2020	-3	-21	-21	-54	9.5	-28	-9.4	-23	-25	-52	11	-31
	2013-2017	-41	-45	-18	-62	37	-20	-36	-44	-17	-63	24	-18
Shijiazhuang	2015-2020	-33	-30	-15	-75	31	-32	-37	-32	-14	-71	26	-32
	2018-2020	-17	-21	-7.4	-40	-5.2	-18	-21	-22	-6.6	-31	-0.8	-20
	2013-2017	-33	-0.8	19	-41	45	4.3	-37	-0.02	8.4	-30	42	-0.6
Shanghai	2015-2020	-41	-44	-18	-61	-5.1	-23	-38	-38	-24	-57	4	-21
	2018-2020	-11	-21	-8.3	-31	-3.5	1.3	-11	-19	-20	-26	4	-2.9
	2013-2017	-42	-36	-8.2	-53	48	4	-45	-38	-14	-54	46	-2.1
Nanjing	2015-2020	-44	-42	-28	-63	11	-20	-40	-36	-25	-58	12	-20
	2018-2020	-26	-27	-15	-29	1.7	7.8	-21	-21	-14	-24	6.6	4.7
	2013-2017	-28	-17	-11	-57	26	-6.2	-33	-17	-14	-58	22	-12
Hangzhou	2015-2020	-45	-32	-15	-61	-4.4	-19	-43	-31	-15	-60	-1.4	-17
	2018-2020	-21	-16	-2.8	-34	-4.5	-17	-20	-15	-5.2	-32	1.2	-17
	2013-2017	-29	-0.8	2.7	-43	-10	-8.7	-32	-4.4	-5.3	-44	-17	-10
Guangzhou	2015-2020	-40	-27	-18	-42	25	-22	-36	-23	-15	-40	22	-20
	2018-2020	-32	-18	-21	-24	5.8	-4.1	-29	-18	-19	-24	5.8	-2.5
	2013-2017	-25	-15	-15	-27	23	-31	-24	-15	-18	-26	25	-30
Shenzhen	2015-2020	-34	-29	-31	-27	-0.1	-33	-25	-18	-29	-23	11	-31
	2018-2020	-29	-21	-22	-15	-9.1	-8.8	-22	-14	-17	-14	-1	-7
	2013-2017	-32	-16	47	-40	60	29	-29	-8.5	38	-41	61	14
Lanzhou	2015-2020	-27	-27	0.6	-24	25	-28	-20	-17	6.8	-14	18	-21
	2018-2020	-18	-28	3.2	-15	-3.1	-3.8	-12	-21	5.3	-6.2	-9	0
	2013-2017	-39	-25	-13	-70	-5.6	-4.7	-45	-31	-18	-71	2.4	-12
Wuhan	2015-2020	-46	-45	-25	-54	7.4	-21	-43	-45	-27	-52	12	-16
	2018-2020	-20	-23	-16	-1.6	3.5	-12	-19	-23	-18	1	9.8	-11
	2013-2017	-37	-38	-10	-57	15	-17	-39	-37	-9.7	-54	26	-21
Chengdu	2015-2020	-33	-38	-25	-62	-1.9	-32	-34	-38	-27	-61	2.2	-34
	2018-2020	-16	-19	-14	-34	-11	-15	-13	-17	-17	-32	-7.1	-13
	2013-2017	-24	-17	39	-64	18	1.6	-30	-22	32	-64	22	-2.2
Chongqing	2015-2020	-39	-37	-10	-51	14	-27	38	-34	-7.7	-48	13	-26
	2018-2020	-14	-14	-6.9	-9.6	-7.8	-11	-14	-14	-6.9	-7.7	1	-10
	2013-2017	-34	-25	0	-56	24	-9	-34	-25	-3.4	-56	21	-11
Average in 12 cities	2015-2020	-39	-37	-19	-59	11	-29	-36	-34	-19	-55	13	-27
	2018-2020	-19	-22	-13	-29	-1.9	-12	-17	-20	-14	-25	2.1	-12