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2	Assessing the impact of Clean Air Action on Air Quality Trends in
3 4	Beijing Wiegacity using a machine learning technique
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24 ABSTRACT

25 A five-year Clean Air Action Plan was implemented in 2013 to reduce air pollutant emissions and 26 improve ambient air quality in Beijing. Assessments of this Action Plan is an essential part of the 27 decision-making process to review the efficacy of the Plan and to develop new policies. Both 28 statistical and chemical transport modelling have been previosuly applied to assess the efficacy of 29 this Action Plan. However, inherent uncertainties in these methods mean that new and independent 30 methods are required to support the assessment process. Here, we applied a machine learning-31 based random forest technique to quantify the effectiveness of Beijing's Action Plan by decoupling 32 the impact of meteorology on ambient air quality. Our results demonstrate that meteorological 33 conditions have an important impact on the year to year variations in ambient air quality. Further 34 analysis show that thePM_{2.5} mass concentration would have broken the target of the Plan (2017 annual $PM_{2.5} < 60 \ \mu g \ m^{-3}$) were it not for the meteorological conditions in winter 2017 favouring 35 36 the dispersion of air pollutants. However, over the whole period (2013 to 2017), the primary emission controls required by the Action Plan have led to significant reductions in PM_{2.5}, PM₁₀, 37 38 NO₂, SO₂ and CO from 2013 to 2017 of approximately 34%, 24%, 17%, 68%, and 33%, 39 respectively, after meteorological correction. The marked decrease in PM_{2.5} and SO₂ is largely 40 attributable to a reduction in coal combustion. Our results indicate that the Action Plan has been 41 highly effective in reducing the primary pollution emissions and improving air quality in Beijing. 42 The Action Plan offers a successful example for developing air quality policies in other regions of 43 China and other developing countries.

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45 Keywords: Clean air action plan, Beijing, air quality, emission control, coal combustion

46 1. INTRODUCTION

47 In recent decades, China has achieved rapid economic growth and become the world's second 48 largest economy. However, it has paid a high price in the form of serious air pollution problems 49 caused by the rapid industrialization and urbanization associated with its fast economic growth 50 (Lelieveld et al., 2015; Zhang et al., 2012; Guan et al., 2016). According to the World Bank, air 51 pollution costs China's economy \$159 billion (~9.9 % of GDP equivalent) in welfare losses and 52 was associated with 1.6 million deaths in China in 2013 (Xia et al., 2016; World Bank and IHME, 53 2016). Accordingly, air pollution has been receiving much attention from both the public and 54 policymakers in China, especially in Beijing - the capital of China with around 22 million 55 inhabitants- which has suffered extremely high levels of air pollutants (Rohde and Muller, 2015; 56 Guo et al., 2013; Zhu et al., 2012; Cai et al., 2017). To tackle air pollution problems, China's State 57 Council released the action plan in 2013 which set new targets to reduce the concentration of air 58 pollutants across China (CSC, 2013). Within the plan, a series of policies, control and action plans 59 with a focus on Beijing-Tianjin-Heibei, the Yangtze River Delta and the Pearl River Delta regions 60 were proposed. To implement the national Action Plan and further improve air quality, Beijing 61 Municipal Government (BMG) formulated and released the "Beijing 2013-2017 Clean Air Action 62 Plan" (the "Action Plan"), which set a target for the mean concentration of fine particles ($PM_{2.5}$, particulate matter with aerodynamic diameter less than 2.5 μ m) to be below 60 μ g m⁻³ by 2017 63 (BMG, 2013). Since then, the five-year period of 2013-2017 has seen the implementation of 64 65 numerous regulations and policies in Beijing.

It is of great interest to the government, policymakers and the general public to know whether the Action Plan is working to meet the set targets. Research in this area is often termed as an air quality accountability study (HEI, 2003; Henneman et al., 2017; Cheng et al., 2018). This is highly challenging because both the actions taken to reduce the air pollutants and the meteorological conditions affect the air quality levels during a particular period (Henneman et al., 2017; Cheng et
al., 2018; Liu et al., 2017; Grange et al., 2018; Chen et al., 2019). Therefore, it is essential to
decouple the meteorological impact from ambient air quality data to see the real benefits in air
quality by different actions.

74 Chemical transport models are used widely to evaluate the response of air quality to emission 75 control policies (Wang et al., 2014; Daskalakis et al., 2016; Souri et al., 2016; Chen et al., 2019). 76 However, there are major uncertainties in emission inventories and in the models themselves, 77 which inevitably affect the outputs of chemical transport models (Li et al., 2017; Gao et al., 2018). 78 Statistical analysis of ambient air quality data is another commonly used method to decouple the 79 meteorological effects on air quality (Henneman et al., 2017; Liang et al., 2015), including the 80 Kolmogorov-Zurbenko (KZ) filter model and deep neural networks (Wise and Comrie, 2005; 81 Comrie, 1997; Eskridge et al., 1997; Hogrefe et al., 2003; Gardner and Dorling, 2001). Among 82 these models, the deep neural network models showed a better performance (i.e., higher correlation 83 coefficient, lower root mean square error - RMSE) but did not allow us to investigate the effect 84 of input variables (therefore it is referred as a "black- box" model) (Gardner and Dorling, 2001; 85 Henneman et al., 2015). More recently, new approaches based on regression decision trees are 86 being developed, which are suitable for air quality weather detrending, including the boosted 87 regression trees (BRT) and random forest (RF) algorithms (Carslaw and Taylor, 2009; Grange et 88 al., 2018). These machine learning based techniques have a better performance than the traditional 89 statistical and air quality models by reducing variance/bias and error in high dimensional data sets 90 (Grange et al., 2018). However, similar to the deep learning algorithms including neural networks, 91 it is hard to interpret the working mechanism inside these models as well as the results. In addition, 92 the decision trees models are prone to over-fitting, especially when the number of tree nodes is

93 large (Kotsiantis, 2013). An over-fitting problem of a random forest model is checked by its ability
94 to reproduce observations using an unseen training data set. Recently published R-packages can
95 partly explain and visualise random forest models including the importance of input variables and
96 their interactions (Liaw and Wiener, 2018; Paluszynska, 2017).

97 Here, we applied a machine learning technique based upon the random forest algorithm and the 98 latest R-packages to quantify the role of meteorological conditions in air quality and thus evaluate 99 the effectiveness of the Action Plan in reducing air pollution levels in Beijing. The results were 100 compared with the latest emission inventory as well as results from previous study which used a 101 chemical transport model - the Weather Research and Forecasting (WRF)-Community Multiscale 102 Air Quality (CMAQ) model (Wong et al., 2012; Xiu and Pleim, 2001).

103 **2.** M

MATERIALS AND METHODS

104 2.1 Data Sources Hourly air quality data for six key air pollutants (PM_{2.5}, PM₁₀, NO₂, SO₂, O₃, 105 and CO) was collected by 12 national air quality monitoring stations in Beijing by the China 106 National Environmental Monitoring Network (CNEM). Hourly air quality data were downloaded 107 from the CNEM website - http://106.37.208.233:20035. Since air quality data are removed from 108 the website on a daily basis, data were automatically downloaded to a local computer and 109 combined to form the whole dataset for this paper. All data are now available at 110 https://github.com/tuanvvu/Air_Quality_Trend_Analysis (last access 5 June 2019). These sites 111 were classified in three categories (urban, suburban, and rural areas). The map and categories of 112 the monitoring sites are given in Figure S1 and Table S1. Hourly meteorological data including wind speed (ws), wind direction (wd), temperature, relative humidity (RH) and pressure recorded 113 114 at Beijing International Airport were downloaded using the "worldMet"- R package (Carslaw,

2017b). Monthly emissions of air pollutants were from the Multi-resolution Emission Inventory
for China (<u>http://www.meicmodel.org/</u>), and for the whole Beijing region. Data was analyzed in
R Studio with a series of packages, including the "openair", "normalweatherr", and
"randomForestExplainer" (Liaw and Wiener, 2018; Carslaw and Ropkins, 2012; Carslaw, 2017a;
Paluszynska, 2017).

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2.2 Random forest modelling

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Figure 1 shows a conceptual diagram of the data modelling and analysis which consists of threesteps:

124 1) Building the random forest (RF) model

A decision tree-based random forest regression model describes the relationships between hourly concentrations of an air pollutant and their predictor features (including time variables: month 1 to 12, day of the year from 1 to 365, hour of a day from 0 to 23, and meteorological parameters: wind speed, wind direction, temperature, pressure, and relative humidity). The RF regression model is an ensemble-model which consists of hundreds of individual decision tree models. The RF model is described in detail in Breiman (1996 & 2001).

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In the RF model, the bagging algorithm, which uses bootstrap aggregating, randomly samples observations and their predictor features with replacement from a training data set. In our study, a single regression decision tree is grown in different decision rules based on the best fitting between the observed concentrations of a pollutant (response variable) and their predictor features. The predictor features are selected randomly to gives the best split for each tree node. The hourly predicted concentrations of a pollutant are given by the final decision as the outcome of the weighted average of all individual decision tree. By averaging all predictions from bootstrap
samples, the bagging process decreases variance, thus helping the model to minimize over-fitting.

As shown in Figure 1, the whole data sets were randomly divided into: 1) a training data set to construct the random forest model and 2) a testing data set to test the model performance with unseen data sets. The training data set comprised of 70% of the whole data, with the rest as testing data. The RF model was constructed using R-"normalweatherr" packages by Grange et al. (2018).

The original data sets contain hourly concentrations of air pollutants (response) and their predictor features that include time variables (t_{trend} - Unix epoch time, the day of the year, week/weekend, hour) and meteorological parameters (wind speed, wind direction, pressure, temperature, and relative humidity). These time predictor features represent effects upon concentrations of air pollutants by diurnal, weekday/weekend day and seasonal cycles and t_{trend} (Unix epoch time) represents the trend in time which captures the long-term change of air pollutant due to changes in policies/regulations, which was calculated as:

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$$t_{trend} = year_i + \frac{t_{JD}-1}{N_i} + \frac{t_H}{24N_i}$$

where, N_i is the number of days in a year i (the year ith from 2013 to 2017), t_H: diurnal hour time (0-23); t_{JD}: day of the year (1-365)) (Carslaw and Taylor, 2009).

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Table S2, Figure S3-S4 and Section S3 provided information on the performance of our model to
reproduce observations based on a number of statistical measures including mean square error
(MSE)/ root mean square error (RMSE), correlation coefficients (r²), FAC2 (fraction of predictions
with a factor of two), MB (mean bias), MGE (mean gross error), NMB (normalised mean bias),

161 NMGE (normalised mean gross error), COE (Coefficient of Efficiency), IOA (Index of 162 Agreement) as suggested in a number of recent papers (Emery et al. 2017, Henneman et al., 2017, 163 and Dennis et al., 2010. These results confirm that the model performs very well in comparison 164 with traditional statistical methods and air quality models (Henneman at al., 2015).

- 165
- 166 **2) Weather normalisation using the RF model**

167 A weather normalisation technique predicts the concentration of an air pollutant at a specific 168 measured time point (e.g., 09:00 on 01/01/2015) with randomly selected meteorological 169 conditions. This technique was firstly introduced by Grange et al. (2018). In their method, a new 170 dataset of input predictor features including time variables (day of the year, the day of the week, 171 hour of the day, but not the Unix time variable) and meteorological parameters (wind speed, wind 172 direction, temperature and RH) is firstly generated (i.e., re-sampled) randomly from the original 173 observation dataset. For example, for a particular day (e.g., 01/01/2011), the model randomly 174 selects the time variables (excluding Unix time) and weather parameters at any day from the data 175 set of predictor features during the whole study period. This is repeated 1,000 times to provide the 176 new input data set for a particular day. The input data set is then fed to the random forest model 177 to predict the concentration of a pollutant at a particular day (Grange et al., 2018; Grange and 178 Carslaw, 2019). This gives a total of 1,000 predicted concentrations for that day. The final 179 concentration of that pollutant, referred hereafter as weather normalised concentration, is 180 calculated by averaging the 1000 predicted concentrations. This method normalises the impact of 181 both seasonal and weather variations. Therefore, it is unable to investigate the seasonal variation 182 of trends for a comparison with the trend of primary emissions. For this reason, we enhanced the 183 meteorological normalisation procedure.

185 In our algorithm, we firstly generated a new input data set of predictor features, which includes 186 original time variables and re-sampled weather data (wind speed, wind direction, temperature, and 187 relative humidity). Specifically, weather variables at a specific selected hour of a particular day 188 in the input data sets were generated by randomly selecting from the observed weather data (i.e., 189 1988-2017 or 2013-2017) at that particular hour of different dates within a four-week period (i.e., 190 2 weeks before and 2 weeks after that selected date). For example, the new input weather data at 191 $08:00\ 15/01/2015$ are randomly selected from the observed data at 08:00 am on any date from 1st to 29th January of any year in 1988-2017 or 2013-2017. The selection process was repeated 192 193 automatically 1,000 times to generate a final input data set. Each of the 1,000 data was then fed to 194 the random forest model to predict the concentration of a pollutant. The 1,000 predicted 195 concentrations were then averaged to calculate the final weather normalised concentration for that 196 particular hour, day, and year. This way, unlike Grange et al., (2018), we only normalise the 197 weather conditions but not the seasonal and diurnal variations. Furthermore, we are able to re-198 sample observed weather data for a longer period (for example, 1998-2017), rather than only the 199 study period. This new approach enables us investigate the seasonality of weather normalised 200 concentrations and compare them with primary emissions from inventories.

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202 3) Quantifying long-term trend using Theil-Sen estimator

The Theil-Sen regression technique was performed on the concentrations of air pollutants after meteorological normalisation to investigate the long-term trend of pollutants. The Theil-Sen approach which computes the slopes of all possible pairs of pollutant concentrations and takes the median value, has been commonly used for long-term trend analysis over recent years. By selecting the median of the slopes, the Theil-Sen estimator tends to give us accurate confidence
intervals even with non-normal data and non-constant error variance (Sen, 1968). The Theil-Sen
function is provided via the "openair" package in R.

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211 **2.3.** Notices, regulations and policies for air pollution control in Beijing

212 The five-year period of 2013-2017 saw the implementation of numerous regulations and policies. 213 The "Beijing Clean Air Action Plan 2013-2017" proposed eight key regulations including: (1) 214 Controlling the city development intensity, population size, vehicle ownership, and environmental 215 resources, (2) Restructuring energy by reducing coal consumption, supplying clean and green 216 energy, and improving energy efficiency, (3) promoting public transport, implementing stricter 217 emission standards, eliminating old vehicles and encouraging new and clean energy vehicles, (4) 218 Optimizing industrial structure by eliminating polluting capacities, closing small polluting 219 enterprises, building eco-industrial parks and pursuing cleaner production, (5) Strengthening 220 treatment of air pollutants and tightening environmental protection standards, (6) Strengthening 221 urban management and regulation enforcement, (7) Preserving the ecological environment by 222 enhancing green coverage and water area, and (8) Strengthening emergency response to heavy air 223 pollution. We collected more than 70 major notices and policies on air pollution control from the 224 Beijing government website (http://zhengce.beijing.gov.cn/library/). Most important regulations 225 were related to energy system re-structuring and vehicle emissions (Section S2). These key 226 measures include: 1) Reform and upgrade Action Plan for coal energy conservation and emission 227 reduction (2014); 2) "no-coal zone" for Beijing-Tianjin-Hebei regions in October 2014; 3) Beijing 228 implemented the fifth phase emission standards for new light-duty gasoline vehicles (LDVs) and

229	heavy-duty diesel vehicles (HDVs) for public transport in 2013; 4) traffic restrictions to yellow-
230	label and non-local vehicles to enter the city within the sixth ring road during daytime since 2015.
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3. **RESULTS AND DISCUSSIONS**

233 **3.1 Observed Levels of Air Pollution in Beijing During 2013-2017**

234 The annual mean concentration of PM_{2.5} and PM₁₀ in Beijing measured from the 12 national air quality monitoring stations declined by 34 and 19 % from 88 and 110 μ g m⁻³ in 2013 to 58 and 89 235 μg m⁻³ in 2017, respectively. Similarly, the annual mean levels of NO₂ and CO decreased by 16 236 and 33 % from 54 μ g m⁻³ and 1.4 mg m⁻³ to 45 μ g m⁻³ and 0.9 mg m⁻³ while the annual mean 237 concentration of SO₂ showed a dramatic drop by 68 % from 23 μ g m⁻³ in 2013 to 8.0 μ g m⁻³ in 238 239 2017. Along with the decrease of annual mean concentration, the number of haze days (defined as $PM_{2.5} > 75 \ \mu g \ m^{-3}$ here) also decreased (Figure S7). These results confirm a significant 240 241 improvement of air quality and that Beijing appeared to have achieved its PM_{2.5} target under the Action Plan (annual average PM_{2.5} target for Beijing is 60 μ g m⁻³ in 2017). On the other hand, the 242 243 annual mean concentration of $PM_{2.5}$ is still substantially higher than China's national ambient air quality standard (NAAQS-II) of 35 µg m⁻³ (Table S3) and the WHO Guideline of 10 µg m⁻³. While 244 245 PM₁₀, PM_{2.5}, SO₂, NO₂ and CO showed a decreasing trend, the annual average concentration of O_3 increased slightly by 4.9 % from 58 µg m⁻³ in 2013 to 61 µg m⁻³ in 2017. The number of days 246 247 exceeding NAAQS-II standards for O₃-8h averages (160 μ g m⁻³) during the period 2013-2017 was 248 329, accounting for 18 % of total days.

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3.2 Air Quality Trends After Weather Normalisation

251 A key aspect in evaluating the effectiveness of air quality policies is to quantify separately the 252 impact of emission reduction and meteorological conditions on air quality (Carslaw and Taylor, 253 2009;Henneman et al., 2017), as these are the key factors regulating air quality. By applying a 254 random forest algorithm, we showed the normalised air quality parameters, under the 30-year 255 average (1988-2017) meteorological conditions (Figure 2). The temporal variations of ambient 256 concentrations of monthly average $PM_{2.5}$, PM_{10} , CO, and NO₂ do not show a smooth trend from 257 2013 to 2017 because of the spikes during pollution events. However, after the weather 258 normalisation, we can clearly see the decreasing real trend (Figure 2). The trends of the normalised 259 air quality parameters represent the effects of emission control and, in some cases, associated 260 chemical processes (for example, for ozone, $PM_{2.5}$, PM_{10}). SO₂ showed a dramatic decrease while 261 ozone increased year by year (Figure 2). The normalised annual average levels of PM_{2.5}, PM₁₀, SO₂, NO₂, and CO decreased by 7.4, 7.6, 3.1, 2.5, and 94 µg m⁻³ year⁻¹, respectively, whereas the 262 level of O_3 increased by 1.0 µg m⁻³ year⁻¹. 263

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265 Table 1 compares the trends of air pollutants before and after normalisation, which are largely 266 different depending on meteorological conditions. For example, the annual average concentration of fine particles (PM_{2.5}) after weather normalisation was 61 μ g m⁻³ in 2017, which was higher than 267 268 their observed level of 58 μ g m⁻³ by 5.2%. This suggests that Beijing would have missed its PM_{2.5} target of 60 μ g m⁻³ if not for the favorable meteorological conditions in winter 2017 and the 269 emission reduction contributed to 10 μ g m⁻³ out of the 13 μ g m⁻³ (77%) PM_{2.5} reduction (71 to 58 270 271 μ g m⁻³) from 2016 to 2017. Overall, the emission control led to a 34%, 24%, 17%, 68%, and 33% reduction in normalised mass concentration of PM2.5, PM10, NO2, SO2 and CO respectively from 272 273 2013 to 2017 (Table 1).

When meteorological conditions were randomly selected from 2013-2017 (instead of 1998-2017) in the RF model, the normalised level of $PM_{2.5}$ in 2017 was 60 µg m⁻³, which is 1 µg m⁻³ difference to that using 1998-2017 data. This difference is due to the variation of the long-term climatology (1998-2017) to the 5 year period (2013-2017)

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The observed $PM_{2.5}$ mass concentration reduced by 30 µg m⁻³ from 2013 to 2017, whereas the normalised values reduced by 32 µg m⁻³. Similarly, the observed PM_{10} and SO_2 mass concentration reduced by 30 and 15.5 µg m⁻³ from 2013 to 2017, whereas the normalised values were 33 and 17.9 µg m⁻³. These results suggest that the effect of emission reduction would have contributed to an even better improvement in air quality (except ozone) from 2013 to 2017 if not for meteorological variations year by year.

Figure 3 shows that the Action Plan has been led to a major improvement in the air quality of Beijing at both the urban, suburban and rural sites, particularly for SO₂ (16-18 % year⁻¹), CO (8-9 % year⁻¹), and PM_{2.5} (6-8 % year⁻¹). The Action Plan also led to a decrease in PM₁₀ and NO₂ but to a lesser extent than that of CO, SO₂ and PM_{2.5}, indicating that PM₁₀ and NO₂ were affected by other less well controlled sources or different atmospheric processes. Urban sites showed a bigger decrease in PM_{2.5}, PM₁₀, and SO₂ concentrations in comparison to the rural and suburban sites (Figure 3).

3.3 Impact of Meteorological Conditions on PM_{2.5} levels: A Comparison with Results from CMAQ-WRF Model

We compared our RF modelling results with those from an independent method by Cheng et al. (2018) who evaluated the de-weathered trend by simulating the monthly average $PM_{2.5}$ mass concentrations in 2017 by the CMAQ model with meteorological conditions of 2013, 2016 and 2017 from the WRF model. The WRF-CMAQ results predict that the annual average $PM_{2.5}$ 2017 from the WRF model. The WRF-CMAQ results predict that the annual average $PM_{2.5}$ 2018 concentration of Beijing in 2017 is 61.8 and 62.4 µg m⁻³ under the 2013 and 2016 meteorological 2019 conditions respectively, both of which are higher than the measured value – 58 µg m⁻³. Thus, the 300 modelled results are similar to those from the machine learning technique, which gave a weather-301 normalised $PM_{2.5}$ mass concentration of 61 µg m⁻³ in 2017.

Figure 4 also shows that the $PM_{2.5}$ concentrations would have been significantly higher in November and December 2017 if under the meteorological conditions of 2016. In contrast, the $PM_{2.5}$ concentrations would have been lower in spring 2017 under the meteorological conditions of 2016 or the 30-year normalised meteorological data. The more favourable meteorological conditions in the two months contributed appreciably to the lower measured annual average $PM_{2.5}$ level in 2017. It also suggests that the monthly levels of $PM_{2.5}$ strongly depend upon the monthly variation of weather.

309 Comparison of model uncertainties from the two methods

310 Figure 5 compares observation and prediction of monthly concentrations of PM_{2.5} by the WRF-CMAQ model and the RF model. The correlation coefficient r^2 between monthly values was 0.82, 311 312 whereas that from the random forest method is >0.99 for both the training and test data sets. The 313 difference between the monthly observed PM_{2.5} values and those simulated by the WRF-CMAO 314 model ranged from 3 to 33.6%, resulting in 7.8% difference in the yearly value. In contrast, the 315 deviation between observed and predicted PM_{2.5} value from the RF model ranges from 0.4-7.9% 316 with an average of 1.5%. In the modelled concentration of $PM_{2.5}$ from the random forest technique, 317 Standard deviation of the 1,000 predicted concentration of PM_{2.5} in 2017 is only 0.35 μ g m⁻³, 318 accounting for 0.6% of the observed PM_{2.5} concentration.

320 3.4 Evaluating the Effectiveness of the Mitigation Measures in the Clean Air Action 321 Plan

322 The weather normalised air quality trend (Figure 2) allows us to assess the effectiveness of various 323 policy measures to improve air quality to some extent. In particular, the SO₂ normalised trend 324 clearly shows that the peak monthly concentration in the winter months decreased from 60 µg m⁻ ³ in January 2013 to less than 10 µg m⁻³ in December 2017 (Figure 2). This indicates that the 325 326 control of emissions from winter-specific sources was highly successful in reducing SO₂ 327 concentrations. The Multi-resolution Emission Inventory for China (MEIC) shows a major 328 decrease in SO₂ emissions from heating (both industrial and centralized heating) and residential 329 sector (mainly coal combustion) (Figure S8), which is consistent with the trend analyses. On the 330 other hand, the "baseline" SO₂ concentration –defined as the minimum monthly concentration in 331 the summer (Figure 2) – also reduced somewhat during the same period. SO_2 in the summer mainly 332 came from non-seasonal sources including power plants, industry, and transportation (Figure S9). 333 Overall, the MEIC estimated that SO₂ emissions decreased by 71 % from 2013 to 2017 (Figure 334 S8), which is close to the 67% decrease in the weather normalised concentration of SO_2 (Table 1). 335 According to the Beijing Statistical Year Books (2012-2017), coal consumption in Beijing 336 declined remarkably by 56 % in 6 years as shown in Figure 6 (Karplus et al., 2018; BMBS, 2013-337 2017). The slightly faster decrease in SO₂ concentrations relative to coal consumption (Figure S9) 338 was attributed to the adoption of clean coal technologies that were enforced by the "Action Plan 339 for Transformation and Upgrading of Coal Energy Conservation and Emission Reduction (2014-340 2020)" (Karplus et al., 2018; Chang et al., 2016). In summary, energy re-structuring, e.g., 341 replacement of coal with natural gas (Figure 6; Section S2), is a highly effective measure in 342 reducing ambient SO₂ pollution in Beijing.

344 Coal combustion is not only a major source of SO₂, but also an important source of NO_x and 345 primary particulate matter (PM) in Beijing (Streets and Waldhoff, 2000; Zíková et al., 2016; Lu et 346 al., 2013; Huang et al., 2014). Precursor gases including SO_2 and NO_x from coal combustion also 347 contribute to secondary aerosol formation (Lang et al., 2017). The MEIC emission inventory 348 showed that 8.8-29 % of NO_x was emitted from heating, power and residential activities, primarily 349 associated with coal combustion. As shown in Figure S9, the normalised NO₂ concentration is also 350 decreasing, but much slower than that of SO_2 . Most notably, the level of SO_2 dropped rapidly in 351 2014 but the level of NO₂ decrease by a small proportion. The different trends between SO₂ and 352 NO₂ indicate that other sources (e.g. traffic emissions, Figure S9) or atmospheric processes have 353 a greater influence on ambient concentration of NO_2 than coal combustion. For examples the 354 chemistry of the NO/NO₂/O₃ system will tend to "buffer" changes in NO₂ causing non-linearity in 355 NO_x-NO₂ relationships (Marr and Harley, 2002). NO₂ concentrations decreased more rapidly from 356 January 2015, specifically by 17%, 18%, 10%, 15% (Figure 2) in the first six months of 2015, 357 which suggests that emission control measures implemented in 2015 were effective. These 358 measures include regulations on spark ignition light vehicles to meet the national fifth phase 359 standard, and expanded traffic restrictions to certain vehicles, including banning entry of high 360 polluting and non-local vehicles to the city within the sixth ring road during daytime, and phasing 361 out of 1 million old vehicles (Yang et al., 2015) (Section S2).

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Normalised $PM_{2.5}$ decreased faster than NO₂, but slower than SO₂ (Figure S9). Yearly peak normalised $PM_{2.5}$ concentrations decreased from 2013-14 to 2015-2016 but slighted rebounded in 2016-2017. The monthly normalised peak $PM_{2.5}$ concentration reduced from 115 µg m⁻³ in Jan

2013 to 60 µg m⁻³ in Dec 2017. The biggest drop is seen in winter 2017, which decreased by more 366 367 than half from the peak value in winter 2016, suggesting that the "no coal zone" policy (Section 368 S2) to reduce pollutant emissions from winter specific sources (i.e., heating and residential sectors) 369 was highly effective in reducing $PM_{2.5}$. The normalised "baseline" concentration – minimum monthly average concentration in the summer – also decreased from 71 μ g m⁻³ in summer 2013 to 370 42 µg m⁻³ in summer 2017. This suggests that non-heating emission sources, including industry, 371 372 industrial heating and power plants also contributed to the decrease in $PM_{2.5}$ from 2013 to 2017. 373 These are broadly consistent with the PM_{2.5} and SO₂ emission trends in MEIC (Figure S8). A small 374 peak in both PM_{2.5} and CO in June/July seen in Figure 2 from 2013 to 2016 attributed to 375 agricultural burning almost disappeared over the period of the measurements and simulations in 376 2017, suggesting the ban on open burning is effective.

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378 The normalised trend of PM₁₀ is similar to that of PM_{2.5}, except that the rate of decrease is slower. 379 The trend agrees well with PM₁₀ primary emissions for the summer (Figure S8). The biggest drop 380 in peak monthly PM_{10} concentration is seen in winter 2017, which decreased by more than half 381 from the peak value in winter 2016, suggesting that "no coal zone" policy (Section S2) to reduce 382 pollutant emission from winter specific sources (i.e., heating and residential sectors) were highly 383 effective in reducing PM₁₀, as with PM_{2.5}. The rate of decrease of peak monthly PM₁₀ emission is 384 slower than that of weather normalised PM₁₀ concentrations, which may suggest an 385 underestimation of the decrease by the MEIC. The normalised "baseline" concentration (minimum 386 monthly average concentration, Figure 2)- also decreased substantially from 2013 to 2017. This 387 indicates that non-heating emission sources, including industry, industrial heating and power 388 plants also contributed to the decrease in PM_{10} . This is consistent with the trends in MEIC (Figure 389 S8). The peaks in the spring are attributed to Asian dust events.

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391 The normalised CO trend shows that the peak CO concentration reduced by approximately 50% 392 from 2013 to 2017 with the largest drop from 2016 to 2017 (Figure 2). The decreasing trend in 393 total emission of CO in the MEIC is slower from 2015 to 2017, suggesting that CO emission in 394 the MEIC may be overestimated in these two years. During 2013-2016, the CO level decreased by 395 26 % and 34 % for winter and summer. Similar to the normalised PM_{2.5} trend, a small peak of CO 396 concentration occurred in Jun-July during 2013-2016, which is likely associated with open 397 biomass burning around the Beijing region. This peak disappeared in 2017. A major decrease in 398 normalised CO levels in winter 2017 is attributed to the "no-coal zone" policy (see below Section 399 S2; Figure S8).

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401 **3.5** Implications and Future Perspectives

402 We have applied a machine learning based model to identify the key mitigation measures 403 contributing to the reduction of air pollutant concentrations in Beijing. However, three challenges 404 remain. Firstly, it is not always straightforward to link a specific mitigation measure to 405 improvement in air quality quantitatively. This is because often more than two measures were 406 implemented on a similar timescale, making it difficult to disentangle the impacts. Secondly, we 407 were not able to compare the calculated benefit for each mitigation measure with that intended by 408 the government due to a lack of information about the implemented policies, for example, the 409 start/end date of air pollution control actions. If data on the intended benefits are known, this will 410 further enhance the value of this type of study. Thirdly, the ozone level increased slightly during 411 2013-2017, especially for the summer periods (Table 1). Because ozone is a secondary pollutant, 412 interpretation of the effects of emission changes of precursor pollutants is complex and beyond the 413 scope of this study.

414

415 Our results confirm that the "Action Plan" has been led to a major improvement in the real 416 (normalised) air quality of Beijing (Figure 3). However, it would have failed to meet the target for 417 annual average PM_{2.5} concentrations if not for better than average air pollutant dispersion 418 (meteorological) conditions in 2017. This suggests that future target setting should consider 419 meteorological conditions. Major challenges remain in reducing the $PM_{2.5}$ levels to below 420 Beijing's own targets, as well as China's national air quality standard and WHO guidelines. 421 Another challenge is to reduce the NO_2 and O_3 levels, which show little decrease or even an 422 increase from 2013 to 2017. The lessons learned in Beijing thus far may prove beneficial to other 423 cities as they develop their own clean air strategies.

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668 669	TABLE LEGENDS:
670 671	Table 1: A comparison of the annual average concentrations of air pollutants before and after weather normalization
672	weather normalisation
673 674	FIGURE I FOENDS.
675	FIGURE LEGENDS.
676	Figure 1: A diagram of long-term trend analysis model
677	Figure 2: Air quality and primary emissions trends
678	Figure 3: Yearly change of air quality in different area of Beijing
679	Figure 4: Relative change in monthly PM _{2.5} levels in 2017 under different weather conditions
680	Figure 5: Comparison of MRF-CMAQ and RF models' performance
681	Figure 6: Primary energy consumption in Beijing
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706	Table 1. A comparison of the annual average concentrations of air pollutants before and after
707	weather normalisation.

Pollutants	PM2.5		PM10		NO ₂		SO ₂		CO		O 3	
year	Obs.	Model	Obs.	Model	Obs.	Model	Obs.	Model	Obs.	Model	Obs.	Model
2013	88	93	110	123	54	58	23	26.3	1.4	1.5	58	59
2014	84	85	119	121	57	56	20	20	1.2	1.3	55	56
2015	80	75	107	106	50	50	13	13	1.3	1.2	58	59
2016	71	71	98	101	47	48	10	10	1.1	1.1	63	60
2017	58	61	90	93	45	48	7.5	8.4	0.9	1.0	60	61

Note: Obs: observed concentration. Model.: Modelled concentration of a pollutant after weather normalisation. Unit: μg m⁻³ for all pollutants, except CO (mg m⁻³) 710





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Figure 2. Air quality and primary emissions trends. Trends of monthly average air quality 739 parameters before and after normalisation of weather conditions (first vertical axis), and the 740 primary emissions from the MEIC inventory (secondary vertical axis). "Model" in the figure means the modelled concentration of a pollutant after weather normalisation. The red line shows 741 742 the Theil-Sen trend after weather normalisation. The black and blue dot lines represent weather 743 normalised and ambient (observed) concentration of air pollutants. The red dot line represents total 744 primary emissions. The levels of air pollutants after removing the weather's effects decreased significantly with median slopes of 7.2, 5.0, 3.5, 2.4, and 120 μ g m⁻³ year⁻¹ for PM_{2.5}, PM₁₀, SO₂, 745 NO₂, and CO, respectively, while the level of O₃ slightly increased by 1.5 μ g m⁻³ year⁻¹. 746

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751 Figure 3. Yearly change of air quality in different area of Beijing. This figure presents yearly average changes of weather normalised air pollutant concentrations at rural, suburban and urban sites (see Figure S1 for classification) of Beijing from 2013 to 2017. Specifically, average yearly changes are for SO₂ (-14%, -15%, -16% year⁻¹- for rural, suburban, and urban areas, respectively), CO (-9%, -9%, -8% year⁻¹), PM_{2.5} (-7%, -8%, -9% year⁻¹), PM₁₀ (-6%, -5%, -7% year⁻¹), NO₂ (-2%, -6%, -5% year⁻¹) and O₃ (1%, 0.3%, 2% year⁻¹). The error on the bar shows the minimum and maximum yearly change.



Figure 4. Relative change in monthly PM_{2.5} levels in 2017 under different weather conditions. This figures presents relative changes (%) in monthly average modelled PM_{2.5} concentrations in 2017 if under the 2016 (red) and 2013 (green) meteorological condition using CMAQ model and under averaged 30 years of meteorological condition using the machine learning technique. A positive value indicates PM_{2.5} concentration would have been higher in 2017 if under the 2013 or 2016 meteorological conditions. Under the meteorological condition of 2016, monthly PM_{2.5} concentration in 2017 would have been approximately 28% lower in January but 53% to 82% higher in November and December. This suggests that 2017 meteorological conditions were very favourable for better air quality comparing to those in 2016. If under the meteorological condition of 2013, monthly PM_{2.5} concentration in 2017 would have been higher in January (22%) and February (36%) but only slightly higher in November (12%) and December (14%).



Figure 5. Comparison of predicted monthly average PM_{2.5} mass concentrations by the WRF CMAQ (Cheng et al., 2018) and RF model against observations in Beijing. WRF-CMAQ results
 are averaged over the whole Beijing region and the observed values refer to the average
 concentration of PM_{2.5} over the 12 sites.



Figure 6. Primary energy consumption in Beijing. Petroleum consumption remained stable (21-23 million tonnes coal equivalent (Mtce)) over the years while natural gas and primary electric power increased significantly by 1.8 times and reached 23 Mtce in 2016. Coal consumption declined remarkably by 56.4% from 15.7 Mtce in 2013 to 6.8 Mtce in 2016. The proportion of coal in primary energy consumption in 2016 was 9.8%, within its target of 10% set by the Beijing government.