



# Measurement Report: A Multi-Year Study on the Impacts of Chinese New Year Celebrations on Air Quality in Beijing, China.

3

4 Benjamin Foreback<sup>1,2</sup>, Lubna Dada<sup>2</sup>, Kaspar Dällenbach<sup>2</sup>, Chao Yan<sup>1,2</sup>, Lili Wang<sup>3</sup>, Biwu Chu<sup>2,4</sup>, Ying Zhou<sup>1</sup>,

5 Tom V. Kokkonen<sup>2,6</sup>, Mona Kurppa<sup>7</sup>, Rosaria E. Pileci<sup>5</sup>, Yonghong Wang<sup>2</sup>, Tommy Chan<sup>2</sup>, Juha

6 Kangasluoma<sup>1,2</sup>, Lin Zhuohui<sup>1</sup>, Yishou Guo<sup>1</sup>, Chang Li<sup>1</sup>, Rima Baalbaki<sup>2</sup>, Joni Kujansuu<sup>1,2</sup>, Xiaolong Fan<sup>1</sup>,

7 Zemin Feng<sup>1</sup>, Pekka Rantala<sup>2</sup>, Shahzad Gani<sup>2</sup>, Federico Bianchi<sup>1,2,</sup>, Veli-Matti Kerminen<sup>2</sup>, Tuukka Petäjä<sup>1,2,6</sup>,

- 8 Markku Kulmala<sup>1,2,6</sup>, Yongchun Liu<sup>1</sup> and Pauli Paasonen<sup>1,2</sup>
- 9
- 10 <sup>1</sup>Aerosol and Haze Laboratory, Beijing Advanced Innovation Center for Soft Matter Science and Engineering,
- 11 Beijing University of Chemical Technology, Beijing, China
- 12 <sup>2</sup>Institute for Atmospheric and Earth System Research / Physics, Faculty of Science, University of Helsinki,
- 13 Finland
- 14 <sup>3</sup>Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China
- <sup>4</sup> State Key Joint Laboratory of Environment Simulation and Pollution Control, Research Center for Eco-
- 16 Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
- 17 <sup>5</sup>Laboratory of Atmospheric Chemistry, Paul Scherrer Institute (PSI), 5232 Villigen, Switzerland
- 18 <sup>6</sup>Joint International Research Laboratory of Atmospheric and Earth System Sciences, School of Atmospheric
- 19 Sciences, Nanjing University, Nanjing, China
- 20 <sup>7</sup>Atmospheric Composition Research, Finnish Meteorological Institute, Helsinki, Finland
- 21
- 22 Corresponding Author: Pauli Paasonen (pauli.paasonen@helsinki.fi)
- 23
- 24

# 25 ABSTRACT

26 We investigated the influence of the Chinese New Year (CNY) celebrations on local air quality in Beijing from 27 2013 through 2019, bringing together comprehensive observations at the newly-constructed Aerosol and Haze 28 Laboratory at Beijing University of Chemical Technology - West Campus (BUCT-AHL) and data from 29 Chinese government air quality measurement stations. In this study, these datasets are used together to provide 30 a detailed analysis of air quality during the CNY over multiple years. Before CNY in 2018, the city of Beijing prohibited the use of fireworks and firecrackers in an effort to reduce air pollution. In 2018 air pollutant 31 concentrations still showed a significant peak during the CNY night, even though not as strong as in previous 32 33 years, but in 2019, the pollution levels were notably lower. During the studied 7-year study period, it appears 34 that there has been a long-term decrease in CNY related emissions since 2016. Based on our analysis, the 35 pollutants with the most notable spike during CNY were sulfur dioxide and particulate matter, including black 36 carbon. Sulfuric acid concentration followed the sulfur dioxide concentration and showed elevated overnight 37 concentration in CNY 2018, but not notably in 2019. Additionally, spectrometer data and analysis of aerosol 38 particle number size distribution shows direct emissions of particles with diameters around 20 nm during CNY 39 in 2018 and 2019. Meteorological conditions were comparable between the latest two years, indicating that air 40 quality associated with the CNY may be improving, perhaps a positive effect of the restrictions. The longer observations in the future will provide confirmation for these trends. 41

42

#### 43 1 INTRODUCTION

44

45 Anthropogenic emissions associated with festivities, notably fireworks and firecrackers (hereafter simply 46 fireworks), are known for their hazardous effects, and even short-term exposure can have significant impacts

47 on human health (Bach et al., 2007; Chen et al. 2011; Jiang et al. 2015; Yang et al. 2014). Firework celebrations





48 are found to increase the concentrations of trace gases and particle concentrations (Kong et al. 2015; Li et al. 49 2013). Furthermore, some studies have related these festivities to the occurrence of haze episodes in the days 50 following a firework event (Li et al. 2013; Feng et al. 2012).

51

52 The CNY is a traditional annual holiday occurring in wintertime - in January or February (the exact date is 53 based on the lunar cycle). Because of the adverse impacts on health, pollution from fireworks during the CNY 54 has gathered attention worldwide. For instance, studies including Yang et al. (2014) in Jinan, Shi et al. (2014) in Tianjin, and Feng et al. (2012) and Zhang et al. (2010) in Shanghai have shown that there is noticeable 55 56 degradation in air quality associated with Chinese New Year celebrations in these cities. The effects of 57 fireworks on air pollution are known for various holidays in other countries as well. Studies in India, for 58 example, during the country's annual Diwali Festival in the late autumn have also shown results of high 59 pollution from firework use (Ravindra et al. 2003; Mönkkönen et al. 2004; Barman et al. 2007; Singh et al. 60 2009; Yerramesetti et al. 2013). As another example, a study by Liu et al. (1997) in Southern California, USA 61 showed enhanced concentrations of particulate matter and trace gas pollutants during firework celebrations.

62

Beginning in 2018, a prohibition on firework burning within the 5<sup>th</sup> Ring Road of Beijing was implemented
(Liu et al. 2019). The study by Liu et al. (2019) reported that the prohibition resulted in about a 40% decrease
in the total number of fireworks and firecrackers sold in Beijing around the 2018 CNY compared to 2016.
Furthermore, the amount of toxic pollution during the 2018 CNY was significantly less than that in 2016.

67

68 In this study, we focus on the measurements collected from the Beijing University of Chemical Technology, 69 Aerosol and Haze Laboratory (BUCT-AHL, Liu et al., 2020), an academic research station in Beijing China, 70 along with seven years of data from the Chinese Ministry of Environmental Protection (MEP) throughout the 71 Beijing metropolitan area. The long-term datasets also provide spatial context in the scale of the greater Beijing 72 area, including a comparison of measurements inside versus outside of the prohibition area. Here we 73 investigated years 2013-2019. The 2020 CNY has not been included in this study because of the widespread 74 impacts of the COVID-19 virus that affected China during this time. Due to the unfortunate circumstance, many 75 Chinese citizens refrained from travel, public celebrations, and time spent in public. Consequently, the 2020 76 CNY is not directly comparable to previous years.

77

78 The aim of this this paper is to provide a detailed view on how CNY celebrations have influenced air quality, 79 atmospheric chemistry and gas-to-particle conversion in Beijing. We start with an in-depth analysis of data from 2018 and 2019 while the longer, 7-year data set provides the perspective into the impacts of the imposed 80 81 restrictions on firework use in the Beijing area. The specific questions we aim to answer include: i) how do the CNY celebrations and associated increase in precursor and aerosol emissions reflect into the atmospheric 82 83 concentrations of trace gases and particulate matter and particle number size distribution; ii) how are these 84 changes connected with meteorological conditions; iii) how does the influence of CNY to regional air quality 85 vary spatially over the Beijing area; iv) how the influence of CNY on Beijing air quality has changed during 86 the recent years, including the result of the firework prohibition beginning in 2018; and v) how does the gas 87 phase sulfuric acid relate to the new particle formation and cluster mode particle number concentration during 88 CNY. Our insights are useful for scientists and policy makers around the world who are interested in improving 89 air quality during holidays that involve firework celebrations. Improving air quality, even short-term, could 90 have a significant positive impact on health and wellbeing of citizens.

91

# 92 2 METHODS

93 94 95





- Data collected for this study have been measured at the newly constructed station near the third ring road of
  Beijing (39°56'N, 116°17'E; Figure 1, Liu et al., 2020). The station, known as the Aerosol and Haze Laboratory,
  is located on Beijing University of Chemical Technology West Campus, which is a five-floor building nearby
- by to a busy highway. The station, hereafter BUCT-AHL, is following the concept of the Station for Measuring
- 100 Ecosystem-Atmosphere Relations (SMEAR) in Hyytiälä, Southern Finland (Hari and Kulmala, 2005). BUCT-
- 101 AHL was built in collaboration with the Institute of Atmospheric and Earth System Research (INAR) at the
- 102 University of Helsinki as part of the effort to build a global SMEAR network (Kulmala, 2018), with the aim to
- 103 understand atmospheric chemical cocktail in megacity (Kulmala, 2015).
- 104
- 105 In our analysis the following datasets from BUCT-AHL during the 2018 and 2019 CNY are used: 1) Trace gas
- 106 concentrations: nitrogen oxides (NO<sub>X</sub>), sulfur dioxide (SO<sub>2</sub>), ozone (O<sub>3</sub>), and carbon monoxide (CO), 2) PM2.5
- 107 aerosol mass concentration, 3) Black carbon mass concentration (BC), 4) Sub-micron aerosol particle number
- 108 size distributions, 5) Gas-phase sulfuric acid ( $H_2SO_4$ ) concentration, 6) Meteorological observations.
- 109

110 Additionally, datasets from several national air quality monitoring sites (NAQMS; Song et al. 2017; Tao et al.

111 2016) within Beijing obtained from the Chinese Ministry of Environmental Protection (MEP) were utilized as 112 follows: 1) Fine and coarse particulate matter mass concentrations (PM2.5 and PM10), 2) trace gases (NOX, 113 SO2, O3, and CO) from 2013 through 2019 for a multi-year comparison. This also provided insights into the 114 spatial variability within the Beijing city and particularly contrasting the area, where the ban for the fireworks

spatial variability within the Beijing city and particularly contrasting the area, where the ban for the firew was implemented against the urban background air quality.

116

#### 117 2.2 Instrumentation

118

#### 119 2.2.1 Observations in BUCT-AHL station

120

#### 121 Trace gas measurements

122 Concentrations of carbon monoxide (CO), sulfur dioxide ( $SO_2$ ), ozone ( $O_3$ ) and nitrogen oxides ( $NO_x$ ) were 123 measured with Thermo Environmental Instruments models 48i, 43i-TLE, 42i, and 49i, respectively. They were 124 all sampled through a common inlet through the roof of the building. The length of the sampling tube was 125 approximately 3 m long (Zhou et al. 2020). The time resolution of the measurements was 5 minutes, but to be 126 consistent with the MEP datasets, one hour averages were used in this study.

127

#### 128 Meteorological observations

Meteorological datasets were collected with a Vaisala automatic weather station, AWS310 on the rooftop of
 BUCT-AHL including wind speed and direction, ambient air temperature and relative humidity. Boundary
 layer height was measured using a Vaisala CL-51 ceilometer on the rooftop of BUCT-AHL.

132

#### 133 Sub-micron aerosol particle number size distributions and total number concentrations

Particle size distribution between 3 nm and 1 $\mu$ m was measured using a particle size distribution (PSD) (Liu et al., 2016). The PSD is composed of a nano-scanning mobility particle sizer (nano-SMPS, 3–55 nm, mobility diameter), a long SMPS (25–650 nm, mobility diameter) and an aerodynamic particle sizer (APS, 0.55–10  $\mu$ m, aerodynamic diameter). It was fitted with a cyclone to remove particles larger than 10  $\mu$ m from entering the system. Sampling was done from the rooftop using a 3 m long sampling tube. Additional information about the setup of these instruments can be found in Zhou et al. (2020).

140

141 Particle sizes have been further divided into four modes, based on particle diameter: cluster mode (sub-3 nm),

- nucleation mode (3–25 nm), Aitken mode (25–100 nm), and accumulation mode (100–1000 nm). The method of is described in Zhou et al. (2020)
- 143 of is described in Zhou et al. (2020).





144

Furthermore, at BUCT-AHL, aerosol particle number size distribution of aerosol particles from 2.5 to 42 nm
 was measured with a neutral cluster and air ion spectrometer (NAIS; model 4-11, Airel, Estonia; Manninen et
 al., 2016; Mirme and Mirme, 2013). The NAIS sampled outside air from a horizontally oriented, 4 cm diameter

- copper sampling tube extending 1.6 m out of a north-facing window. The sampling flow rate was 54 l min<sup>-1</sup>
  (Zhou et al., 2020).
- 150

Additionally, an Airmodus A11 Nano Condensation Nucleus Counter (nCNC) system, commonly known as PSM (Vanhanen et al., 2011) was used to measure the sub 3nm particle number concentration. The PSM was operated in scanning mode in which the saturator flow is continuously ramped between 0.1 and 1.3 lpm and back to 0.1 lpm. The sampling line was 1.2 m long and having the same orientation as the NAIS sampling line.

### 155

#### 156 Gas-phase sulfuric acid

157 Sulfuric acid was measured by a chemical ionization atmospheric-pressure interface time-of-flight mass 158 spectrometer equipped with a nitrate chemical ionization source (CI-APi-TOF, Jokinen et al., 2012). The 159 ionization was done with NO3- as the reagent ion in ambient pressure (e.g., Petäjä et al., 2009). Nitrate reagent 160 ion was produced by photo-ionizing a mixture of 3 mL.min<sup>-1</sup> ultrahigh purity nitrogen flow containing nitric acid with 20 mL.min<sup>-1</sup> zero air. This mixture acted as the sheath flow and was introduced into a coaxial laminar 161 162 flow reactor concentric to the sample flow. The sample flow was 8.8 L min<sup>-1</sup> but only 0.8 L.min<sup>-1</sup> was drawn 163 into the pinhole of the TOF. The sampling line was 1.6 m long stainless-steel tube having an inner diameter of 3/4 inch and positioned horizontally. The instrument was calibrated with known concentrations of sulfuric acid. 164 165 Further information about the calibration procedure can be found in Kürten et al. 2012.

166

#### 167 Black carbon mass concentration

An aethalometer AE33 (Magee Scientific) monitored the light absorption related to the aerosol. Equivalent
 black carbon (eBC) was computed based on the change in time of the light attenuation using procedures
 presented in Virkkula et al. (2015)

171

#### 172 2.2.2 Chinese MEP Data

Beginning in 2013, the Chinese Ministry of Environmental Protection began installing sensors China-wide to
measure local, regional, and large-scale air quality. Real-time datasets from this sensor network are published
hourly by the China Environmental Monitoring Center (CEMC), which includes PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>2</sub>, NO<sub>X</sub> and
CO. There are over 1000 active sensors across China (Song et al. 2017; Tao et al. 2016).

177

In this study, data from 12 MEP sites throughout Beijing are used (See Table 1 in Supplementary Information
for a list of these sites and their locations). The Guanyuan (GY) site is the closest site to BUCT-AHL, about 5
km east. The original data are available at http://106.37.208.233:20035/ and for this study we have removed

181 the outliers with criteria presented by Wu et al. (2018).

#### 182 3 RESULTS AND DISCUSSION

183

184 Effects of elevated pollutant emissions during the Chinese New Year were observed both at BUCT-AHL and 185 the MEP sites during the analysis periods. Effects include sudden spikes in concentrations of trace gases, 186 particles, and BC. We explore the time series of the observations in the section below in more detail.

187

#### 188 3.1 Characteristics of air quality during the Chinese New Year





Figure 2 shows a timeseries of air pollutant concentrations from eight days before to eight days after the 2018 and 2019 CNY at BUCT-AHL. The CNY was on February 16, 2018 and February 5, 2019. In the BUCT measurements, we observed sharp peaks in Particulate Matter mass (PM<sub>2.5</sub>), SO<sub>2</sub>, sulfuric acid, CO, BC, NO, and NO<sub>2</sub> and ozone during firework events. These observations agree with the previous studies showing a

194 connection between holiday-related firework celebrations and degraded air quality (Jiang et al., 2015; Yang et

- al., 2014; Shi et al., 2014; Feng et al., 2012; Zhang et al., 2010).
- 196

197 Figure 2 shows that in 2018, a significant spike in PM<sub>2.5</sub> concentration is observed overnight, in particular at

midnight, on the CNY. Additionally, in 2018 period of haze for three days following the CNY was observed.

199 In contrast, in 2019,  $PM_{2.5}$  was observed to have lower concentration on the CNY night than on the previous

and following nights. There is also a noticeable spike in SO<sub>2</sub> overnight of the CNY in 2018, shown in Figure

- 201 3, but less noticeable alteration of  $SO_2$  is observed overnight of the 2019 CNY.
- 202

The measurements show clearly elevated nighttime concentration of  $H_2SO_4$  on CNY in 2018, concentration exceeding  $3*10^6$  cm<sup>-3</sup> during the whole night, whereas on other nights such high concentrations are observed only occasionally. In 2019, there is no signs of anomalies in nighttime  $H_2SO_4$  concentration during CNY. An unknown spike in  $H_2SO_4$  is noticed at noon the day before CNY in 2018, and its association with celebratory activities is unclear. Like with  $PM_{2.5}$  and  $SO_2$ , Figure 2 shows a clearly distinctive spike in BC around midnight of the 2018 CNY. However, there appears to be little to no effect of CNY on BC in 2019.

209

The measurements show elevated concentration of  $NO_2$  overnight of the CNY in both years, yet no spike in NO concentration. Fireworks emit  $NO_2$  but not NO (Jiang et al., 2015); however, a high  $NO_2/NO_x$  ratio can also be caused by accumulation of pollutants emitted the previous afternoon (Chou et al., 2009). Nonetheless, when comparing the CNY characteristics of  $NO_x$  with other pollutants, there is a noticeable spike of  $NO_2$  during the CNY during both years.

215

216 Generally, Figure 2 shows that during the CNY celebrations in 2018 concentrations of all the primary pollutants 217 are elevated, implying enhanced direct emissions. The concentrations of sulfuric acid and ozone react to 218 elevated concentrations as expected, sulfuric acid concentration increases due to enhanced formation with 219 increased SO<sub>2</sub> concentration, and ozone concentration decreases with increased chemical sink by NO<sub>x</sub> and CO 220 (and probably other carbon compounds). However, on 2019, only the concentrations of CO and  $NO_2$  are 221 observed to increase during CNY celebrations, leading to decrease in ozone concentration, whereas the 222 concentrations of all other pollutants did not show elevated concentrations. Interestingly, the measurements in 223 Figure 2 show degraded air quality between 16-20 February 2018 immediately following the Chinese New 224 Year, which closely resembles the characteristics of a haze event as described in Zhao et al. (2013) and Zhao 225 et al. (2011). These haze events have elevated concentrations of pollution continuously for multiple days, and 226 these concentrations gradually increase throughout the episodes, and they end with sudden decline, often caused 227 by a cold front or synoptic weather system. Several previous studies, including Jiang et al. (2015) and Li et al. 228 (2013), suggest that fireworks likely contribute to haze formation. Therefore, the increased level of pollutants 229 observed overnight during the 2018 CNY have likely contributed to this subsequent haze period. However, the 230 meteorological conditions are also important for haze formation, which are discussed in Section 3.2.

231

#### 232 **3.2 Effects of Meteorology and Boundary Layer Height**

233

Because meteorology conditions vary between different years, it is important to understand its effects on pollution when comparing different years to each other. Most notably, wind, humidity, boundary layer height,

and precipitation can affect pollutant concentrations during and after the fireworks.





238 The wind speed during the night of the 2018 CNY peaks at  $\sim 2$  m/s, whereas during the night of the 2019 CNY, 239 it remains less than  $\sim 1$  m/s. Temperature and relative humidity are quite comparable between the years. 240 Precipitation was not measured at BUCT-AHL in either year. However, online weather data shows there was 241 no precipitation in the region during either of the vears 242 (https://www.wunderground.com/history/weekly/cn/beijing/ZBNY/date/2019-2-4). The nocturnal boundary 243 layer heights are low in both years (Figure 2), which is unfavorable for vertical mixing of pollutants. This, 244 along with the low wind speeds, points towards more efficient dispersion in pollutant concentrations in 2018 245 than in 2019, indicating that the reason for clearly lower pollutant concentrations in 2019 is likely related to 246 lower emissions, not meteorological conditions.

247

#### 248 **3.3** Particle number concentrations and size distribution

249

250 To further explore the effects the fireworks on air pollution, Figure 3 shows particle number concentrations and 251 size distributions measured by the NAIS instrument at BUCT-AHL from the day before to the day after CNY. 252 The results show that shortly before midnight on CNY in 2018, an elevated concentration of aerosol particles 253 with diameters of roughly 20 nm was observed, simultaneous to the spike in  $SO_2$  concentration. This increase 254 in particle number concentration is not associated with a regional new particle formation (NPF) event (e.g. 255 Mäkelä et al., 1997; Shen et al., 2011; Heintzenberg et al. 2007), which in this case is taking place on the 256 following day before noon, where the concentrations of well below 10 nm particles are first observed to increase 257 and then to grow to 20 nm sizes. This feature suggests that the observed particles during festivities are of 258 primary origin and emitted to atmosphere in the respective size range, possibly from the fireworks. Similar to 259 most other pollutants, particle number concentrations in this size range in CNY 2019 do not show signs of 260 celebration related emissions. There is a continuous concentration of particles in the respective size range, but 261 no clear peaks during the celebrations, in line with no signs of increased SO<sub>2</sub> concentration.

262

Aerosol particle mass concentration (PM2.5) during the CNY firework period is clearly elevated reaching values close to 250  $\mu$ g m<sup>-3</sup> and 150  $\mu$ g m<sup>-3</sup> at midnight for the year 2018 and 2019, respectively, which are considerable higher than the mass concentrations on the previous day. The midnight peak in PM mass concentration coincides with atmospheric nanoparticle concentrations and elevated SO<sub>2</sub> in 2018 indicating high emissions of primary aerosol particles and co-emitted sulfur dioxide.

268

269 Figure 4 shows the particle number concentrations in four size modes, namely sub-3 nm cluster mode, 3-25 nm 270 nucleation mode, 25-100 nm Aitken mode, and 100-1000 nm accumulation mode, as a function PM<sub>2.5</sub> 271 concentration measured at BUCT-AHL in 2018 and 2019, for 48 hours before through 48 hours after the CNY. 272 The darker colors mark the nighttime measurements on the CNY (9pm-5am). The night-time mass 273 concentrations are noticeably greater. The mass-to-number concentration comparison follows the same general 274 curve during nighttime as the full time period, and the pattern, including the nighttime observations, is 275 consistent with recent investigation by Zhou et al. (2020). The PM2.5 concentrations during the CNY period in 276 2018 are significantly higher than in other periods. The cluster and nucleation mode concentrations are lower, 277 Aitken mode is on the same level, and accumulation mode concentration is higher during the CNY period. This 278 is reasonable, as accumulation mode particles contribute significantly to PM2.5 mass and constitute the major 279 sink for cluster and nucleation mode particles due to coagulation scavenging of smaller particles by larger ones.

280

In figure 5 we can see how the cluster mode concentrations behaves as a function of sulfuric acid. The high nocturnal sulfuric acid concentration during CNY celebrations in 2018 do not lead to high cluster or nucleation mode concentration. In fact, the particle number concentrations in these modes deviates from the otherwise clear response to sulfuric acid concentrations. The reason for this is visible in the panel for accumulation mode

285 concentration vs sulfuric acid concentration: during the CNY 2018 the high concentrations of accumulation





mode particles correlates with sulfuric acid concentration thus plausibly neglecting the enhanced particle cluster and particle formation rates by enhanced coagulation sink as explained earlier.

288

# 289 3.4 Multi-Year Variation of Chinese New Year Effects in Beijing

290

Although fireworks were formally prohibited within the 5<sup>th</sup> Ring Road of Beijing beginning in 2018, there was
still evidence of fireworks burning either in the city or the immediate vicinity of the city, as measured at BUCTAHL, which is within the prohibition area. Nonetheless, because the initial peak during the 2018 CNY (Figure
is significantly higher, even though it disperses more quickly, it is therefore evident that there was more
initial pollution during this time, whereas the amount of pollution during the 2019 CNY was considerably less,
even though it remained present for longer time period.

297

A longer-term multi-year study can be useful in demonstrating whether or not the policy is effective in reducing firework-related pollution, and if there is an overall decreasing trend of pollution effects from fireworks over multiple years. To investigate this question, it is useful to compare the 2018 and 2019 CNY with previous years in Beijing. Datasets have been analyzed from 12 stations in Beijing from 2013 through 2019.

302

Figure 6 shows that each year, there was a spike in pollution around midnight during the CNY. The highest levels were observed in 2016, and the lowest levels were in 2019. Observations from 2013, 2014, 2015, and 2017 also showed similarly high or higher levels of  $PM_{2.5}$  as in 2018 (unfortunately the 2017 dataset is incomplete and does not extend beyond 00:00 of New Year day). Furthermore, data from 2013, 2014, 2015 and 2018 suggest the presence of haze episodes in the days following the New Year, potentially related to firework burning. An elevated level of pollution for two days after the CNY compared to before the CNY was observed in 2019 as well, even though it was to a lesser extent.

310

311 Data from the CNYs have also been compiled into box plots in Figure 7, depicting the distributions of pollutant 312 concentrations from 6:00 pm on CNY Eve to 6:00 am on the CNY day each year. The highest PM 313 concentrations during this time were in 2016 and have since decreased. Concentrations of NO<sub>2</sub>, SO<sub>2</sub>, and CO 314 all show a similar pattern as PM. It should be noted that in 2017, the data is missing after midnight. The decrease 315 since 2016 agrees with the results obtained by Liu et al. 2019. Since ozone is a secondary product and it reacts 316 with several primary pollutants, its concentration pattern being roughly opposite to those of primary pollutants 317 is as expected. In this aspect, it is notable that in 2019 in addition to primary pollutant concentrations also ozone 318 concentration has decreased from 2017 and 2018.

319

# 320 3.5 Spatial variability based on MEP measurement network data

321

322 A further analysis of the CNY in Beijing is to perform a spatial comparison of the MEP measurements across 323 the Beijing region. This includes comparing the observations inside the  $5^{\text{th}}$  Ring Road, where fireworks were 324 prohibited, to outside the ring. Figure 8 maps the 12 MEP stations in the Beijing region for 2013-2019, showing 325 the relative difference of  $PM_{2.5}$  measurements from 9 pm through 5 am during the night of CNY to the average of measurements  $\pm$  48 hours of the CNY at each site. Figures in Supplementary Information show observations 326 327 of PM<sub>2.5</sub> from the 12 individual MEP sites and the corresponding differences, year-by-year from 2013-2019. 328 Based on Figure 8, we can see significant variation from year to year as to which station measures the highest 329 pollution. It is important to note that the population density is greater closer to city center, and thus the 330 population density could impact the results. However, it is plausible to assume that the relative population 331 density difference between the city center and the surrounding areas do not change dramatically during the few 332 year time period.





334 Figure 8 illustrates that in 2013 and 2014, the enhancement in PM<sub>2.5</sub> concentrations during CNY is greater 335 inside the 5<sup>th</sup> ring than outside. In 2015, the enhancement is much greater at the two northeastern sites (HR and SY). In 2016, the differences vary, with no clear difference inside or outside the 5<sup>th</sup> ring. In 2018, the 336 337 enhancement of PM<sub>2.5</sub> is higher inside the 5<sup>th</sup> ring than outside, except for the SY site to the far northeast, which 338 had significantly high enhancement compared to the other sites. In 2019 the enhancement is overall less inside 339 compared to outside.

340

341 Figure 9 shows differences between the PM<sub>2.5</sub> median of the sites inside the 5<sup>th</sup> Ring Road and the median of 342 the sites outside the  $5^{\text{th}}$  Ring (that is the median of the 8 inside sites minus the median of the outside 4 stations) 343 48 hours before through 48 hours after the CNY for 2013-2019. In most years, there is a greater enhancement of PM<sub>2.5</sub> inside than outside. However, in 2015, the opposite was true. In 2016, the first part of the CNY 344 345 overnight had lower PM<sub>2.5</sub> inside than outside, but that reversed a few hours later. In 2019, there was less PM<sub>2.5</sub> 346 inside than outside throughout the night of CNY.

347

#### 348 4 CONCLUSIONS

349

350 This study confirms that CNY consistently impacts air quality in Beijing year-after-year. These results are 351 consistent with previous studies that have linked the CNY (and other similar holiday celebrations involving 352 firework burning around the world) to degraded air quality both locally and regionally. Our results suggest that 353 the regulations to limit firework use have improved the air quality within the restriction zone inside the fifth 354 ring road in Beijing since 2016.

355

356 Based on our observations at BUCT-AHL station in Beijing, in 2018, we detected clearly higher than typical night-time concentrations of particulate mass (PM2.5), particle number, trace gas and sulfuric acid 357 358 concentrations during the CNY. The increase in sulfuric acid concentration did not lead to observed new 359 particle formation, which is explained by simultaneously increasing condensation and coagulation sinks for 360 clustering vapors and freshly formed particles, respectively. However, we observed appearance of particles 361 with diameters of roughly 20 nm that seemed to be linked to enhanced sulfur dioxide concentrations. Based on the MEP data, the peaks in concentrations of different pollutants were noticeably lower than in the previous 362 years. In 2019, a peak in pollution was observed overnight, but it was significantly lower than in 2018, while 363 meteorological conditions were comparable in both years. The significant year-to-year variability depended 364 365 presumably on the meteorological conditions, on new imposed regulations as well as on the fact that the CNY 366 period is determined with a lunar calendar and therefore the exact CNY period varies from year-to-year. In 367 2013, 2014, and 2015, haze episodes lasting several days were observed immediately following the CNY. In 368 2016 and 2018 a moderate haze episode began one day following the CNY.

369

370 Comparing the level of increase in pollutant concentrations during CNY night inside and outside Beijing 5<sup>th</sup> 371 ring road (firework prohibition area) revealed that in 2019 the increase inside this area was smaller than outside. 372 During most, but not all, of the previous CNYs, the increase in concentration was higher inside than outside. 373 This was also the case in 2018. However, as also in previous years the ratio of inside and outside concentrations 374 during CNY has varied, it is unclear if this is related to efficacy of the emission prohibition or, e.g., to larger 375 scale air-mass movements. As absolute concentrations, our results show a decrease of CNY pollution within 376 the prohibition area since 2016 and especially in 2019. This is in agreement with the previous Liu et al. 2019 377 study, which compared the 2016 and 2018 CNY (before and after the prohibition took effect). 378

379 This long-term analysis, which combines BUCT data with multiple years of Chinese government data, 380 demonstrates the importance of analyzing multiple data sources to determine overall trends, rather than making 381 conclusions based on a single dataset. This also demonstrates the usefulness of long-term measurements.

382 Therefore, we suggest ongoing measurements at both BUCT-AHL and MEP sites into multiple future years.





383

The combination of the BUCT-AHL comprehensive observations together with the spatial variability provided by the MEP sites, we see excellent potential that can be utilized to investigate the changes in a) atmospheric chemistry – like ozone dynamics and sulfuric acid formation; b) atmospheric gas-to-particle conversion; c) boundary layer dynamics and d) air quality. Here we have investigated CNYs as case studies to get better insight how rapid changes in emissions will affect the previous four items.

389

#### 390 ACKNOWLEDGEMENTS

391

392 The work is supported by Academy of Finland via Center of Excellence in Atmospheric Sciences (project no. 393 272041) and European Research Council via ATM-GTP 266 (742206). This research has also received funding 394 from Academy of Finland (project no. 316114 & 315203, 307537), Business Finland via Megasense-project, 395 European Commission via SMart URBan Solutions for air quality, disasters and city growth, (689443), ERA-396 NET-Cofund as well as the Doctoral Programme in Atmospheric Sciences at the University of Helsinki. Partial 397 support from the National Key R&D Program of China (2016YFC0200500), and the National Natural Science 398 Foundation of China (91544231 & 41725020) is acknowledged. The authors also wish to acknowledge the 399 Finnish Centre for Scientific Computing (CSC) – IT Center for Science, Finland, for computational resources.

400

### 401 AUTHOR CONTRIBUTIONS

402

All BUCT affiliated authors, plus KD, BC, YW, TC, and PR contributed to measurement collection at BUCT. LW provided the quality-controlled MEP data. BF, LD, KD, TP, FB and MK conceptualized and conducted the data analysis. TK, MoK, RP, and RB participated in the data analysis. TK and MoK provided the meteorology data. KD, TP, FB, PP and MK supervised the study. BF visualized the data and visualized the data and prepared the manuscript with contributions from all other authors. SG assisted with the data visualization and generating figures.

409

#### 410 COMPETING INTERESTS

- 411
- 412 The authors declare that there are no conflicts of interest in this study.
- 413

#### 414 **REFERENCES**

- 415
- Bach, W., Daniels, A., Dickinson, L., Hertlein, F., Morrows, J., Margolis, S., and Dinh, V. D. (2007). Fireworks
  Pollution and Health. *International Journal of Environmental Studies*. 7,1975:183-192.
- 418
- 419 Barman, S. C., Singh, R., Negi, M. P. S., and Bhargava, S. K. (2007). Ambient air quality of Lucknow City
- 420 (India) during use of fireworks on Diwali Festival. *Environ. Monit. Assess.*, 137:495–504.
- 421
- 422 Chen, B., Kan, H., Chen, R., Jiang, S., and Hong, C. (2011). Air Pollution and Health Studies in China—Policy
- 423 Implications. Journal of the Air & Waste Management Association.65-11:1292-1299.
- 424
- 425 Chou, C. C.-K., Tsai, C.-Y., Shiu, C.-J., Liu, S. C., and Zhu, T. (2009). Measurement of NO<sub>v</sub> during Campaign
- 426 of Air Quality Research in Beijing 2006 (CAREBeijing-2006): Implications for the ozone production efficiency
- 427 of NO<sub>x</sub>. Journal of Geophysical Research: Atmospheres. 114:D00G01.
- 428





- 429 Feng, J., Sun, P., Hu, X., Zhao, W., Wu, M., and Fu, J. (2012). The chemical composition and sources of pm<sub>2.5</sub>
- 430 during the 2009 Chinese new year's holiday in Shanghai. *Atmospheric Research*, 118:435–444.
- 431
- 432 Hari, P. and Kulmala, M. (2005). Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II). Boreal
- 433 Environment Research. 10:315-322.
- 434
- 435 He, H., Li, C., Loughner, C. P., Li, Z., Krotkov, N. A., Yang, K., Wang, W., Zheng, Y., Bao, X., Zhao, G., and
- 436 Dickerson, R. R. (2012). SO2 over central China: Measurements, numerical simulations and the tropospheric
- 437 sulfur budget. *Journal of Geophysical Research*, 117, D00K37.
- 438
- Heintzenberg, J, Wehner, B., and Birmili, W. (2007). 'How to find bananas in the atmospheric aerosol': new
  approach for analyzing atmospheric nucleation and growth events. Tellus B: *Chemical and Physical Meteorology*, 59:2, 273-282.
- 442
- Jiang, Q., Sun, Y. L., Wang, Z., and Yin, Y. (2015). Aerosol composition and sources during the Chinese
  Spring Festival: fireworks, secondary aerosol, and holiday effects. *Atmospheric Chemistry and Physics*, 15:6023–6034.
- 446
- Kong, S. F., Li, L., Li, X. X., Yin, Y., Chen, K., Liu, D. T., Yuan, L., Zhang, Y. J., Shan, Y. P., Ji, Y. Q. (2015)
  The impacts of firework burning at the Chinese Spring Festival on
- 449 air quality: insights of tracers, source evolution and aging processes. Atmospheric Chemistry and 450 Physics.15:2167-2184.
- 451
- 452 Kulmala, M. (2015). Atmospheric Chemistry: China's Chocking Cocktail. Nature Comment.
- 453
- 454 Kulmala, M. (2018). Build a global Earth observatory. *Nature Comment*.
- 455
- Kürten, A., Rondo, L., Ehrhart, S., and Curtius, J. (2012). Calibration of a chemical ionization mass
  spectrometer for the measurement of gaseous sulfuric acid. *The Journal of Physical Chemistry A*. 116:63756386.
- 459
- Li, W., Shi, Z., Yan, C., Yang, L., Dong, C., and Wang, W. (2013). Individual metal-bearing particles in a regional haze caused by firecracker and firework emissions. *Sci. Total Environ*, 443, 464-469.
- 462
- Liu, D.-Y., Rutherford, D., Kinsey, M., and Prather, K. A. (1997). Real-Time Monitoring of Pyrotechnically
   Derived Aerosol Particles in the Troposphere. *Analytical Chemistry*, 69, 1808-1814.
- 465
- Liu, J. Q., Jiang, J. K., Zhang, Q., Deng, J. G., and Hao, J. M. (2016). A spectrometer for measuring particle
  size distributions in the range of 3 nm to 10 μm, *Frontiers of Environmental Science & Engineering*, 10:63–
  72.

- Liu, J., Chen, Y., Chao, S., Cao, H., and Zhang, A. (2019). Levels and health risks of PM2.5-bound toxic metals
   from firework/firecracker burning during festival periods in response to management strategies. *Ecotoxicology*
- 472 and Environmental Safety. 171:406-413.
- 473
- 474 Liu, Y.C., Yan, C., Feng, Z., Zheng, F., Fan, X., Zhang, Y., Li, C., Zhou, Y., Lin, Z., Guo, Y., Zhang, Y., Ma,
- 475 L., Zhou, W., Liu, Z., Dada, L., Dällenbach, K., Kontkanen, J., Cai, R., Chan, T., Chu, B., Du, W., Yao, L.,





Wang, Y., Cai, J., Kangasluoma, J., Kokkonen, T., Kujansuu, J., Rusanen, A., Deng, C., Fu, Y., Yin, R., Li, X.,
Lu, Y., Liu, Y., Lian, C., Yang, D., Wang, W., Ge, M., Wang, Y., Worsnop, D.R., Junninen, H., He, H.,
Kerminen, V.-M., Zheng, J., Wang, L., Jiang, J., Petäjä, T., Bianchi, F. and Kulmala, M. (2020) Continuous
and comprehensive atmospheric observation in Beijing: a station to understand the complex urban atmospheric

480 environment, Big Earth Data 4, 295-321.

481

482 Manninen, H. E., Mirme, S., Mirme, A., Petäjä, T., and Kulmala, M. (2016). How to reliably detect molecular

- 483 clusters and nucleation mode particles with Neutral cluster and Air Ion Spectrometer (NAIS), Journal of
- 484 *Atmospheric Measurement Techniques*, 9:3577-3605, 10.5194/amt-9-3577-2016, 2016.
- 485

Mirme, S., and Mirme, A. (2013) The mathematical principles and design of the NAIS – a spectrometer for the
 measurement of cluster ion and nanometer aerosol size distributions. *Journal of Atmospheric Measurement Techniques*, 6:1061-1071, 2013.

489

Mönkkönen, P., Koponen, I.K., Lehtinen, K.E.J., Uma, R., Srinivasan, D., Hämeri, K., and Kulmala, M. (2004).
Death of nucleation and Aitken mode particles: observations at extreme atmospheric conditions and their theoretical explanation. *Journal of Aerosol Science*. 35:781-787.

493

Peltonen, M. (2017). University of Helsinki builds an air quality measuring station in Beijing. University of
 Helsinki News and Press Releases.

496

Ravindra, K., Mor, S., and Kaushik, C. P. (2003). Short-term variation in air quality associated with firework
events: A case study. *Journal of Environ. Monit*, 5. 260–264.

499

Shen, X. J., Sun, J. Y., Zhang, Y.M., Wehner, B., Nowak, A., Tuch, T., Zhang, X. C., Wang, T. T., Zhou, H.
G., Zhang, X. L., Dong, F., Birmili, W., and Wiedensohler, A. (2011). First long-term study of particle number
size distributions and new particle formations of regional aerosol in the North China Plain. *Atmospheric Chemistry and Physics*, 11:1565-1580.

504

Shi, G.-L., Liu, G.-R., Tian, Y.-Z., Zhou, X.-Y., Peng, X., and Feng, Y.-C. (2014). Chemical characteristic and toxicity assessment of particle associated PAHs for the short-term anthropogenic activity event: During the Chinese new year's festival in 2013. *Science of the Total Environment*, 482-483:8–14.

508

Singh, D. P., Gadi, R., Mandal, T.K., Dixit, C.K., Singh, K., Saud, T., Singh, N., and Gupta, P. K. (2009).
Study of temporal variation in ambient air quality during Diwali festival in India. *Environ. Monit. Assess*, 169:1–13.

512

Song, C., Wu, L., Xie, Y., He, J., Chen, X., Wang, T., Lin, Y., Jin, T., Wang, A., Liu, Y., Dai, Q., Liu, B.,
Wang, Y., and Mao, H. (2017). Air pollution in China: Status and spatiotemporal variations. *Environmental Pollution*. 227:334-347.

- 516
- 517 Tao, M., Chen, L., Li, R., Wang, L., Wang, J., Wang, Z., Tang, G., and Tao, J. (2016). Spatial Oscillation of

the particle pollution in eastern China during winter: Implications for regional air quality and climate.
 *Atmospheric Environment*.144:100-110.





- van der A, R. J., Mijling, B., Ding, J., Koukouli, M. E., Liu, F., Li, Q., Mao, H., and Theys, N. (2017) Cleaning
  up the air: effectiveness of air quality policy for SO<sub>2</sub> and NO<sub>x</sub> emissions in China, *Atmospheric Chemistry and Physics.*, 17:1775-1789.
- 524
- Vanhanen, J., Mikkilä, J., Lehtipalo, K., Sipilä, M., Manninen, H. E., Siivola, E., Petäjä, T., and Kulmala, M.
  (2011). Particle Size Magnifier for Nano-CN Detection. *Aerosol Science and Technology*, 45:533-542, 10.1080/02786826.2010.547889.
- 528
- 529 Virkkula, A., Chi1, X., Ding, A., Shen, Y., Nie, W., Qi, X., Zheng, L., Huang, X., Xie, Y., Wang, J., Petäjä,
- Virkula, A., Chil, A., Ding, A., Sheh, T., Ne, W., Qi, A., Zheng, L., Huang, A., Ale, T., Wang, J., Fetaja,
   T., and Kulmala, M. (2015). On the interpretation of the loading correction of the aethalometer. Atmospheric
   Measurement Techniques. 8:4415–4427.
- 532
- Xue, W., Wang, J., Niu, H., Yang, J., Han, B., Lei, Y., Chen, H., and Jiang, C. (2013). Assessment of air quality
   improvement effect under the National Total Emission Control Program during the Twelfth National Five-Year
   Plan in China. *Atmospheric Environment*. 68:74-81.
- 536
- Wu, H. J., Tang, X., Wang, Z. F., Wu, L., Lu, M. M., Wei, L. F., and Zhu, J. (2018). Probabilistic automatic
  outlier detection for surface air quality measurements from the China National Environmental Monitoring
  Network. *Adv. Atmos. Sci.*, 35(12), 1522–1532.
- 540
- Yang, L., Gao, X., Wang, X., Nie, W., Wang, J., Gao, R., Xu, P., Shou, Y., Zhang, Q., and Wang, W. (2014).
   Impacts of firecracker burning on aerosol chemical characteristics and human health risk levels during the
- 542 Chinese New Year celebration in Jinan, China. *Science of the Total Environment*, 476-477:57–64.
- 544
- Yerramsetti, V. S., Anu Rani Sharma, A. R., Navlur, N. G., Rapolu, V., Chitanya Dhulipala, N. S. K. C., and
  Sinha, P. R. (2013). The impact assessment of Diwali fireworks emissions on the air quality of a tropical urban
- 547 site, Hyderabad, India, during three consecutive years. *Environ. Monit. Assess.*, 185:7309–7325.
- 548
- Zhang, M., Wang, X., Chen, J., Cheng, T., Wang, T., Yang, X., Gong, Y., Geng, F., and Chen, C. (2010).
  Physical characterization of aerosol particles during the Chinese new year's firework events. *Atmospheric*
- 551 Environment, 44:5191–5198.
- 552
- Zhao, X., Zhang, X., Pu, W., Meng, W., Xu, X. (2011). Scattering properties of the atmospheric aerosol in
  Beijing, China. *Atmospheric Research*. 101:799-808.
- 555
- Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W. W., Dong, F., He, D., Shi, Q. F. (2013). Analysis of a winter
   regional haze event and its formation mechanism in the North China Plain. *Atmospheric Chemistry and Physics*.
- 558 13:5685-5696.
- 559
- 560 Zhou, Y., Dada, L., Liu, Y., Fu, Y., Kangasluoma, J., Chan, T., Yan, C., Chu, B., Daellenbach, K. R., Bianchi,
- 561 F., Kokkonen, T. V., Liu, Y., Kujansuu, J., Kerminen, V.-M., Petäjä, T., Wang, L., Jiang, J., and Kulmala, M.
- 562 (2020). Variation of size-segregated particle number concentrations in wintertime Beijing. *Atmospheric*
- 563 *Chemistry and Physics.* 20:1201–1216
- 564





565



- 567
- Figure 1: Location of the BUCT-AHL site within the Beijing metropolitan area. © OpenStreetMap
   contributors, CC BY-SA.
- 570









Figure 2: Major pollutants measured in Beijing during the 2018 and 2019 CNY.







Figure 3: Aerosol number size distribution from NAIS instrument from one day before the CNY through one
day following the CNY in 2018 and 2019, overlain with aerosol mass concentration PM<sub>2.5</sub>, black lines) and
SO<sub>2</sub> (blue lines). A spike of PM<sub>2.5</sub> and SO<sub>2</sub> is observed in both years, but significantly less in 2019. Results
from the NAIS show a corresponding release of particles approximately 11 nm in diameter during the time of
the CNY fireworks.







Figure 4: PM<sub>2.5</sub> number concentration as a function of mass concentration in cluster, nucleation, Aitken, and
 accumulation modes in 2018 and 2019, with comparison of CNY ± 48 hours with measurements from 9pm
 through 5am the night of the CNY. Measurements are from BUCT-AHL.









Figure 5: Number concentration of particles in cluster, nucleation, Aitken, and accumulation modes as a function of sulfuric acid concentration in 2018 and 2019, with comparison of CNY ± 48 hours with measurements from 9pm through 5am the night of the CNY. Measurements are from BUCT-AHL.







589

Figure 6: PM2.5 averaged from 12 MEP sites in Beijing (top) and from only the Guanyuan (GY) site, which is the closest MEP measurement site to BUCT-AHL (bottom), from three days before through three days after the 2013-2019 CNY. The highest peak of pollution during the CNY overnight was in 2016, and the lowest was in 2019.







Figure 7: Boxplots of particulate matter and trace gases between 18:00 and 06:00 on the night of the Chinese
New Year in the years 2013-2019. boxplots show 1<sup>st</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup>, and 99<sup>th</sup> percentiles of the data across the
12 sites during this 12-hour period (13 time points, inclusively).

599







600

Figure 8: The 12 MEP sites mapped in the Beijing metropolitan area, showing the ratio of overnight PM<sub>2.5</sub> observations during the CNY (21:00-05:00) to all data during the period of 48 hours before through 48 hours after the CNY. The red line marks the approximate location of the 5<sup>th</sup> Ring Road. Note that the colorbars in each map are relative to only that year, and the colorbar range is not the same in different years. 2017 is omitted from this figure because data after 00:00 was not available. © Google Earth





