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

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ABSTRACT

We investigated This study investigates the influence of the Chinese New Year (CNY) celebrations on local air quality in Beijing from 2013 through 2019, bringing. CNY celebrations include burning of fireworks and firecrackers, which consequently has a significant short-term impact on local air quality. In this study, we bring together comprehensive observations at the newly-constructed Aerosol and Haze Laboratory at Beijing University of Chemical Technology – West Campus (BUCT-AHL) and datahourly measurements from twelve Chinese government air quality measurement stations. In this study, these throughout the Beijing metropolitan area. These datasets are used together to provide a detailed analysis of air quality during the CNY over multiple years. Before CNY in 2018, during which the city of Beijing prohibited the use of fireworks and firecrackers in an effort to reduce air pollution. In 2018 air pollutant concentrations still showed a significant peak during the CNY night, even though not as strong as in previous years, but in 2019, the pollution levels were notably lower. During the studied 7 year study period, it appears that there has been a long term decrease in CNY related emissions since 2016. Based on our analysis, the pollutants with the most notable spike during CNY were sulfur dioxide and particulate matter, including black carbon. before CNY 2018. Datasets used in this study include particulate matter mass concentrations (PM<sub>2.5</sub> and PM<sub>10</sub>), trace gases (NO<sub>X</sub>, SO<sub>2</sub>, O<sub>3</sub>, and CO) and meteorological variables for 2013-2019, aerosol particle size distributions, and concentrations of sulfuric acid and black carbon for 2018 and 2019. Studying the CNY over several years, which has rarely been done in previous studies, can show trends and effects of societal and policy changes over time, and the results can be applied to study problems and potential solutions of air pollution resulting from holiday celebrations. Our results show that during the 2018 CNY, air pollutant concentrations peaked during the CNY night (for example, PM<sub>2.5</sub> reached a peak around midnight of over 250 μg/cm<sup>3</sup>, compared to values of less than 50 μg/cm<sup>3</sup> earlier in the day). The pollutants with the most notable spikes were sulfur dioxide, particulate matter, and black carbon, which are emitted in burning of firework and firecrackers. Sulfuric acid concentration followed the sulfur dioxide concentration and showed elevated overnight concentration in CNY 2018, but not notably in 2019. Additionally, spectrometer data and analysis. Analysis of aerosol particle number size distribution shows showed direct emissions of particles with diameters around 20100 nm during in relation to firework burning. During the 2019 CNY, the pollution levels were somewhat lower (PM<sub>2.5</sub> peaking to around 150 µg/cm<sup>3</sup>

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at CNY in-compared to values around 100 µg/cm³ earlier in the day) and only minor peaks related to firework burning were observed. During both CNYs 2018 and 2019— secondary aerosol formation in terms of particle growth was observed. Meteorological conditions were comparable between the latestthese two years, indicating suggesting that air quality associated CNY-related emissions were lower in 2019 compared 2018. During the 7-year study period, it appears that there has been a general decrease in CNY-related emissions since 2016. For example, peak in PM25 in 2016 was over 600 µg/cm³, and in the years following, the peak was less each year, with the CNY may be improving, perhapsa peak around 150 µg/cm³ in 2019. This is indicative of the restrictions and public awareness of the air quality issues having a positive effect of the restrictions. The longeron improving air quality during the CNY. Going into the future, long-term observations in the future-will provide offer confirmation for these trends.

# 1 INTRODUCTION

Anthropogenic emissions associated with festivities, notably fireworks and firecrackers (hereafter simply fireworks), are known for their hazardous effects, and even short-term exposure can have significant impacts on human health (Bach et al., 2007; Chen et al., 2011; Jiang et al., 2015; Yang et al., 2014). Firework celebrations are found to increase the concentrations of trace gases and particle concentrations (Kong et al., 2015; Li et al., 2013). Furthermore, some studies have related these festivities to the occurrence of haze episodes in the days following a firework event (Li et al., 2013; Feng et al., 2012).

The Chinese New Year (CNY) is a traditional annual holiday occurring in wintertime – in January or in February (as the exact date is based on the lunar cycle). Because of the adverse impacts on health, pollution from fireworks during the CNY 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 degradation in air quality associated with Chinese New Year celebrations in these cities. Wang et al. (2007) has shown that firework celebrations emit significant amounts of sulfur dioxide and black carbon. The effects of fireworks on air pollution are known for various holidays in other countries as well. Studies in India, for example, during the country's annual Diwali Festival in the late autumn have also shown results of high pollution from firework use (Ravindra et al., 2003; Mönkkönen et al., 2004; Barman et al., 2007; Singh et al., 2009; Yerramesetti et al., 2013). As another example, a study by Liu et al. (1997) in Southern California, USA showed enhanced concentrations of particulate matter and trace gas pollutants during firework celebrations.

Beginning in 2018, Because of the rising awareness of air quality problems during holiday celebrations, the government of Beijing decided to implement a prohibition on firework burning within the 5<sup>th</sup> Ring Road of Beijing was implemented (Liu et al. 2019). The, in an effort to reduce air pollution, described in a study by Liu et al. (2019)). Their study reported that the prohibition resulted in about a 40% decrease in the total number of fireworks and firecrackers sold in the city of Beijing aroundduring the 2018 CNY holiday compared to 2016. Furthermore, the amount of toxic pollutionLiu et al. (2019) reported that observed concentrations of air pollutants during the 2018 CNY was significantly less than that in 2016.

Therefore, an aim of this study is to confirm the conclusions of Liu et al. (2019) study using not only a 2016 vs. 2018 comparison, but a longer study of each year between 2013 and 2019. Furthermore, this study offers a spatial comparison of the area where fireworks were prohibited (inside the 5th ring) with a region where there was no prohibition (outside the ring). Currently, there are no previous studies that perform such a side-by-side comparison of areas with different firework burning policies.

This manuscript provides a detailed view on how CNY celebrations have influenced air quality and atmospheric chemistry in the Beijing metropolitan area. We start with an in-depth analysis of data from 2018 and 2019, and

then we expand with the longer 7-year dataset. Combined, these datasets provide perspective into the impacts of the imposed restrictions on firework use in the Beijing area. The specific questions we aim to answer include: 1.) how the CNY celebrations and associated increase in precursor and aerosol emissions reflect in the atmospheric concentrations of trace gases and particulate matter and particle number size distribution; 2.) how these changes are connected with meteorological conditions; 3.) how the influence of CNY affects regional air quality variation spatially over the Beijing area; and 4.) how the influence of CNY on Beijing air quality has changed during the recent years, including the result of the firework prohibition beginning in 2018.

## 2 METHODS

InThe observations used in this study, we focus on the include measurements collected from the Beijing University of Chemical Technology, Aerosol and Haze Laboratory (BUCT-AHL, Liu et al., 2020), an academic research station in Beijing China; (Liu et al., 2020); along with seven years of data from twelve measurement stations throughout the Beijing metropolitan area, operated by the Chinese Ministry of Environmental Protection (MEP) throughout the Beijing metropolitan area.). The long-term datasets also provide spatial context in the scale of the greater Beijing area, including a comparison of measurements inside versus outside of the prohibition area. Here we investigated years 2013-2019. The Although data from the 2020 CNY hasis available, we have decided not been included to include it in this study because of the widespread impacts of the COVID-19 virus that affected China during this time. Due to the unfortunate circumstance, many Chinese citizens refrained from travel, public celebrations; and time spent in public. Consequently, the 2020 CNY is not directly comparable to previous years.

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The aim of this this paperThis study is novel and unique in a few ways. First, it is one of the only studies to provide a detailed view on hownot only show measurements for a single CNY celebrations have influenced air quality, atmospheric chemistry and gas to particle conversion in Beijing. We start with an in depth analysis of(or similar celebratory holidays in other countries), but it studies the holiday over seven continuous years. This offers the ability to show trends and effects of, for example policy changes, over time. Furthermore, this study uses data from 2018 and 2019 while the longer, 7 year data set provides the perspective into the impacts of the imposed restrictions on firework use in themultiple institutions, which demonstrates the value of collaborations between different institutions when it comes to solving major global problems such as air pollution. This study also compares the CNY inside the centre of the city to the greater 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 concentrations of trace gases and particulate matter and particle number size distribution; ii) how are these changes connected with meteorological conditions; iii) how does the influence of, which is unique compared to any previous CNY to regional(or similar holiday) air quality vary spatially over the Beijing area; iv) how the influence of CNY on Beijing air quality has changed during the recent years, including the result of the firework prohibition beginning in 2018; and v) how does the gas phase sulfuric acid relate to the new particle formation and cluster mode particle number concentration during CNY.study that uses data at a single location. Our insights are useful foroffer value to scientists and policy makers around the world who are interested in improving air quality during holidays that involve firework celebrations. Improving air quality, even short-term, eouldcan have a significant positive impact on health and wellbeing of citizens.

# 21METHODS

# 2.1 Measurement sites

Data This study uses data collected for this study have been measured atfrom two sources. First, we used data from the newly constructed station near the third ring road of Beijing (39°56'N, 116°17'E; Figure 17; Liu et al., 2020). The station, known as the Aerosol and Haze Laboratory, is located onat Beijing University of Chemical Technology West Campus, which ison the roof of a five-floor building nearby to a busy highway. The station, hereafter (BUCT-AHL, is following) follows the concept of the Station for Measuring Ecosystem-Atmosphere Relations (SMEAR) in Hyytiälä, Southern Finland (Hari and Kulmala, 2005). BUCT-AHL was built in collaboration with the Institute of Atmospheric and Earth System Research (INAR) at the University of Helsinki as part of the effort to build a global SMEAR network (Kulmala, 2018), with the aim to understand atmospheric chemical cocktail in megacity (Kulmala, 2015). In addition to collecting data for in-depth air quality analysis, this joint work increases collaboration between atmospheric scientists in China and Finland.

In our analysis, the following datasets from BUCT-AHL during the 2018 and 2019 CNY are used: 1) Trace gas concentrations: nitrogen oxides (NO<sub>X</sub>), sulfur dioxide (SO<sub>2</sub>), ozone (O<sub>3</sub>), and carbon monoxide (CO<sub>7</sub>); 2) PM2.5 aerosol mass concentration, 3) Black carbon mass concentration (BC), 4); 3) Sub-micron aerosol particle number size distributions, 5; 4) Gas-phase sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) concentration, 6; 5) Meteorological observations. Technical details of the instruments, including manufacturer, parameters measured, time resolution, and available time periods of measurements, can be found in Table S2 in Supplementary material. These details are also described in Liu et al. (2020).

Additionally, we obtained datasets from several national air quality monitoring sites within the Beijing metropolitan area (NAQMS; Song et al., 2017; Tao et al., 2016) within Beijing.). These datasets were obtained from the Chinese Ministry of Environmental Protection (MEP) were utilized as follows), which contain the following: 1) Fine and coarse particulate matter mass concentrations (PM<sub>2.5</sub> and PM<sub>10</sub>), 2) trace gases (NO<sub>X</sub>, SO<sub>2</sub>, O<sub>3</sub>, and CO) from 2013 through 2019 for a multi-year comparison. This also provided insights into the spatial variability within the Beijing city and particularly contrasting the area; where the ban for the fireworks was implemented against the urban background air quality.

#### 2.2 Instrumentation

# 2.2.1 Observations in BUCT-AHL station

# Trace gas measurements

Concentrations of carbon monoxide (CO), sulfur dioxide (SO<sub>2</sub>), ozone (O<sub>3</sub>) and nitrogen oxides (NO<sub>x</sub>) were measured with Thermo Environmental Instruments models 48i, 43i-TLE, 42i, and 49i, respectively. They were all sampled through a common inlet through the roof of the building. The length of the sampling tube was approximately 3 m long (Zhou et al., 2020). The time resolution of the measurements was 5 minutes, but to be consistent with the MEP datasets, one—hour averages were used in this study.

# Meteorological observations

Meteorological datasets for 2018-2019 at BUCT-AHL 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 (BLH) was measured using a Vaisala CL-51 ceilometer. Meteorological and BLH measurements were taken on the rooftop of BUCT-AHL.

Archived meteorological data for Beijing from 2013-2017 was obtained from the Weather Underground website (https://www.wunderground.com/history/daily/cn/beijing/ZBNY/). The station used is the Beijing Nanyuan Airport (ICAO identifier ZBNY), a small airport located between the 4<sup>th</sup> and 5<sup>th</sup> Ring Road, south of Beijing city center. The station is approximately 17 km from BUCT-AHL.

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

Particle size distribution (PSD) between 3 nm and 1µm was measured using a particle size distribution (PSD) an instrument of the same name, PSD (Liu et al., 2016). The PSD instrument 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 µm, aerodynamic diameter). It was fitted with a cyclone to remove particles larger than 10 µm 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).

Particle Aerosol 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).

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 1 min<sup>+</sup> (Zhou et al., 2020).

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.

## Gas-phase sulfuric acid

 Sulfuric acid was measured by a chemical ionization atmospheric-pressure interface time-of-flight mass spectrometer equipped with a nitrate chemical ionization source (CI-APi-TOF, Jokinen et al., 2012). The ionization was done with NO<sub>3</sub>- as the reagent ion in ambient pressure (e.g., Petäjä et al., 2009). Nitrate reagent ion wasions were 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<sub>-</sub> with an X-ray source. This mixture acted as the sheath flow and was introduced into a coaxial laminar 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 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. Further information about the calibration procedure can be found in Kürten et al. (2012-).

## 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).

#### 2.2.2 Chinese MEP Data

Beginning in 2013, the Chinese Ministry of Environmental Protection (MEP) began installing sensorsa Chinawide network of air quality monitoring stations to measure local, and regional, and large scale air quality. Real-time datasets from this sensor network are published hourly by the China Environmental Monitoring Center

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(CEMC), which includes  $PM_{10}$ ,  $PM_{2.5}$ ,  $SO_2$ ,  $NO_X$  and CO. There are over 1000 active sensors across China (Song et al., 2017; Tao et al., 2016).

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

# 2.2.3 Back-trajectories with Hysplit

Back trajectories to the BUCT-station were calculated using Hybrid Single-Particle Lagrangian Integrated Trajectory (Hysplit). This model is developed by National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory and the Australian Bureau of Meteorology Research Centre, and it is one of the most widely-used models to determine the origin of an air mass (Stein et al., 2015). In this work, Hysplit trajectories were calculated for the CNY each year from 2013-2019, with the trajectories arriving between 18:00 and 06:00 local time (UTC+8) during the CNY. This adds value to the analysis in two ways: First, it can show whether the air masses in Beijing originated over other urban areas in China, e.g. the greater Beijing-Tianjin-Hebei (BTH) area, or whether the air mass came from more rural areas, e.g. Inner Mongolia or Mongolia. Additionally, it gives a synoptic overview of the weather conditions leading up to CNY. This in turn provides information on whether the air mass is more stagnant within the BTH area, which would result in higher pollution buildup, or whether it originated farther away, which would mean it would be cleaner from the start (Wang et al., 2010; Chen et al., 2015; Zhu et al., 2020).

#### 3 RESULTS AND DISCUSSION

Effects of Higher atmospheric concentrations due to elevated pollutant emissions during the Chinese New Year were observed both at BUCT-AHL and the MEP sites during the analysis periods. Effects The observed features include sudden spikes in concentrations of trace gases, aerosol particles, and BC. We explore the time series of the These observations agree with the previous studies showing a 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). In the sections below, we will delve into these results, which can broaden scientific understanding of the impacts of firework celebrations on local and regional air quality, especially in the section below in more detail context of a wide metropolitan area over the course of several years.

# 3.1 Characteristics of air quality during the Chinese New Year Years 2018 and 2019

The CNY was on February 16, 2018 and February 5, 2019. 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 (except for PM<sub>2.5</sub>, which is from the nearby MEP sites). 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 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). In 2018 the peak in PM<sub>2.5</sub> was over 250 μg/m³, compared to less than 50 μg/m³ half a day before, and in 2019 the peak of PM<sub>2.5</sub> was over 150 μg/m³ compared to less than 100 μg/m³ earlier in the day. Similar spikes in BC, gas phase sulfuric acid, and trace gas concentrations of several times the values earlier in the day were observed in 2018 as well.

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.

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In contrast, in 2019, PM<sub>2.5</sub> was observed to have lowerless noticeable enhancement in concentration—on the CNY night than on the previous and following nights. There is also. While there was a noticeable spike in SO<sub>2</sub> overnight of the CNY in 2018<sub>7</sub> (a spike over 20 ppb compared to less than 5 ppb earlier in the day), shown in Figure 3, but2, a much less noticeable alterationenhancement of SO<sub>2</sub> iswas observed overnight of the 2019 CNY<sub>7</sub> (a peak around 5 ppb compared to around 3 ppb earlier in the day).

The measurements show clearlyshowed elevated nighttime concentration of H<sub>2</sub>SO<sub>4</sub> on CNY in 2018, concentration exceeding 3\*\_10<sup>6</sup> cm<sup>-3</sup> during the whole night, whereas on other nights such high-which was an order of magnitude higher than typical nighttime H<sub>2</sub>SO<sub>4</sub> concentrations are observed only occasionally-of 5·10<sup>5</sup> cm<sup>-3</sup> (Dada et al., 2020). In 2019, there iswas no signsevident indication of anomalies in nighttime H<sub>2</sub>SO<sub>4</sub> concentration during CNY. An unknown spike in H<sub>2</sub>SO<sub>4</sub> iswas noticed at noon the day before CNY in 2018, and its association with celebratory activities is unclear. Like with PM<sub>2.5</sub> and SO<sub>2</sub>, Figure 2 shows a clearly distinctive spike in BC around midnight of the 2018 CNY. However, there appears to be little to no effect These simultaneous peaks of BC and SO<sub>2</sub> during the CNY on BC in 2019 night most likely originate from firework burning.

However, there appeared to be little to no effect of CNY on BC in 2019. The measurements showshowed an elevated concentration of NO₂ overnight of the CNY in both years, (45 ppb in 2018 and 20 ppb in 2019), yet no obvious spike in NO concentration. Fireworks emit NO₂ but not NO (Jiang et al., 2015); however, a∆ high NO₂/NO₃ ratio can also be caused by accumulation of pollutants emitted the previous afternoon (Chou et al., 2009). Nonetheless, when comparing the CNY characteristics of NO₃ with other pollutants, there is a noticeable spike), but in case of NO₂ during the CNY during both years. night it is straight forward to conclude it is due to firework burning, which has been shown to emit NO₂ but no NO (Jiang et al., 2015).

Generally, Figure 2 also shows that during the CNY celebrations in 2018 concentrations of all-the primary pollutants—are-,  $SO_2$ , CO, BC, NO and  $NO_2$ , were elevated, implying enhanced direct emissions-during the CNY period. Secondary pollutants are formed through chemical reactions (Seinfeld and Pandis, 2016) including, for instance, sulfuric acid and ozone. The concentrations of sulfuric acid and ozone react to elevated concentrations these secondary pollutants were as expected. sulfuric acid concentration increases increased due to enhanced formation rate with increased  $SO_2$  concentration, and ozone concentration decreases decreased with increased chemical sink by  $NO_x$  and CO (and probably other carbon compounds). However, onin 2019, only the concentrations of CO and  $NO_2$  are were observed to increase during CNY celebrations, leading to a decrease in ozone concentration, whereas the concentrations of all other pollutants did not show elevated concentrations.

Interestingly, the measurements inin addition to the short-term enhancement of pollutant concentrations. Figure 2 showshows degraded air quality between 16-20 February 2018-immediately, following the Chinese New Year, which closely resembles the characteristics of a haze event as described in Zhao et al. (2013)-and), Zhao et al. (2011)-) and Guo et al. (2020). Using the data from BUCT-AHL, this period was quantifiably classified as a haze event using the algorithm in Zhou et al. (2020). These haze events have elevated concentrations of pollution continuously for multiple days, and these concentrations gradually increase throughout the episodes, and they end. The haze eventually ends with sudden decline, often caused by an arrival of a cold front or change in synoptic weather system conditions. Several previous studies, including Jiang et al. (2015) and Li et al. (2013), suggest that fireworks likely contribute to haze formation. Therefore, It is plausible that the increased level of pollutants observed overnight during the 2018 CNY have likely contributed to this subsequent haze period. However, the meteorological conditions and air mass origins are also important for haze formation, which are discussed in Section 3.2.

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

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Because meteorologythe meteorological conditions during CNY vary between different years, it is important to understand its effects onaddress the impact of local and synoptic scale meteorological parameters on air pollution when comparing different years to each other. Most notably Specifically, wind, speed and direction, relative humidity, (RH), boundary layer height, and precipitation can affect pollutant concentrations during and after the fireworks.

However, none of the measured local meteorological variables showed drastic differences between CNY nights of 2018 and 2019. The wind speed during the night of the 2018 CNY peakspeaked at ~2 m/s, whereas and during the night of the 2019 CNY, it remains remained to values less than ~1 m/s- (Figure 3 and Figure S1 in Supplementary). Temperature and was between 0 and 5 °C on both years. Some difference was observed in relative humidity are quite comparable between the years., as CNY 2018 took place in very dry conditions (RH ~ 20%), whereas during CNY 2019 RH was roughly 40%. Precipitation was not measured at BUCT-AHL in either year. However, online, and weather data showsmeasured at ZBNY show there was no precipitation in the region during either of the www.wunderground.com/history/weekly/cn/beijing/ZBNY/date/2019-2-4). -(data obtained Weather Underground), which was supported by observed RH values below 50%. The nocturnal boundary layer heights are lowwere less than 500 meters in both years (Figure 2), which is unfavorable for vertical mixing of the pollutants. This, along with Due to the lowslightly lower wind speeds, points towards in 2019 than 2018, we would expect more efficient dispersion in pollutants, and thus lower concentrations in 2018. Higher RH is also often related to higher concentrations of aerosol pollutants (Sun et al., 2013). However, what we observed was that there were higher concentrations in 2018 than 2019. This indicates that the reason for lower pollutant concentrations in 2018 than in 2019, indicating that the reason for clearly lower-2019 is not due to differences in the local meteorological conditions.

The lower concentrations in 2019 can be either due to lower emission rates in the area with which the measured air mass is in contact with, or due to a shorter exposure to roughly similar emissions during both years. Figure 4 shows 96 hour back-trajectories by Hysplit, during the night of CNY in 2018 and 2019, showing the sources of the airmasses. This provides further insights into the history of the airmasses in Beijing, including how clean we can expect the airmasses to be before CNY, and whether the airmasses are stagnant around Beijing or whether clean air is being transported into the city. These trajectories show that, prior to CNY midnight, the air mass transport conditions in 2018 and 2019 were rather similar, as the air masses just prior to and during midnight arrived across the high emission areas to the East/South-East/South of Beijing. However, in 2019, the trajectories turned to arrive from West with increasing transport velocity just prior to the midnight. Roughly similar turning is observed in 2018, but instead of occurring some hours before midnight, it occurs after midnight, and the transport velocity from West is smaller than in 2019. This could explain some difference on the pollutant concentrations in 2019 is likely related to lower emissions, not meteorological conditions-concentration levels, but the absence of SO<sub>2</sub> and BC peaks around the CNY midnight in 2019 suggests that local firework burning was not the reason for the elevated PM<sub>2.5</sub> concentration. Overall, our investigation in conditions during the CNY nights of 2018 and 2019 does not offer a clear meteorological explanation to the much lower pollutant concentrations in 2019.

# 3.3 Particle Aerosol particle number concentrations and aerosol number size distribution

To further exploreFurther exploring the effects of the fireworks on air pollution, Figure 35 shows particle number concentrations and size distributions measured by the NAIS instrumentPSD at BUCT-AHL from the day before to the day after CNY. The results show that shortlyShortly before midnight on CNY in 2018, an elevated concentration of aerosol particles with diameters of roughly 20100 nm was observed, simultaneous to

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the spike in SO<sub>2</sub> concentration. This increase in particle After the spike, SO<sub>2</sub> concentration remained elevated until the next morning. PM2.5 concentration increases simultaneously with the SO2 concentration but did not show the same spike than SO<sub>2</sub>, PM<sub>2.5</sub> concentration remained high (>200 µg/m<sup>3</sup>) until the next morning, when it decreased to low values (<30 µg/m³) together with decreasing SO<sub>2</sub> concentration. The nocturnal pollution episode showed a very similar pattern in both SO2 and PM2.5, despite the spike in SO2 occurring together with increased number concentration is not associated with a regional new particle formation (NPF) event (e.g. Mäkelä et al., 1997; Shen et al., 2011; Heintzenberg et al. 2007), which in this case is taking place on the following day before noon, where the concentrations of well below 10 of roughly 100 nm particles are first observed to increase and then to grow to 20 nm sizes. This feature suggests that the observed particles during festivities are of primary origin and emitted to atmosphere in the respective size range, possibly from the fireworks. Similar to most and BC (Figure 2e). This is consistent with air pollution from firework burning. It might have originated from a source nearby, but it can also be transported as a single strong plume from further away, e.g., from outside the 5th Ring Road which was the edge of the prohibited area for firework activity. The overnight elevated concentration of PM2.5 and SO2, excluding the SO2 spike, may be related to accumulated mixture of firework and other pollutants, particle number concentrations in this size range in CNY 2019 do not show signs of celebration festivity related emissions. There is a continuous, e.g., from traffic or cooking. The accumulation of PM25 seems to be related to secondary aerosol formation, since the particle size distribution shows growth of particles in the dominant particle mode during the CNY night (concentration dN/d(log(dp)) over 3.3·10<sup>4</sup> between diameters 40 and 200 nm at around 8 pm and between diameters 60 and close to 300 nm at around 4 am).

In 2019, secondary aerosol mass formation was also observed, as the particle mode grew in diameter steadily between 6 pm and 6 am and the PM<sub>2.5</sub> concentration increased simultaneously until 4 am. The peak PM<sub>2.5</sub> concentration was, however, much lower in 2019 than in 2018 (roughly 100 µg/m³ and close to 250 µm/m³, respectively). SO<sub>2</sub> increased steadily throughout the night and exhibited only a mild peak, from 3 to 6 ppb, shortly after midnight. This peak was again accompanied with simultaneous increase in concentration of particles in the respective size range, but no clear peaks during the celebrations, in line with no signs of increased SO<sub>2</sub> concentration, with diameters around 100 nm and in BC concentration (Figure 2), which suggests contribution from fireworks. However, since the SO<sub>2</sub> concentration showed only a mild peak and did not follow the PM<sub>2.5</sub> concentration, the contribution of nearby firework activity to the overall pollution was estimated to be negligible.

Aerosol particle mass concentration (PM2.5) during the CNY firework period is clearly elevated reaching values close to 250 µg m³ and 150 µg m³ 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.

Figure 46 shows the particle number concentrations in four size modes, namelyspecifically sub-3 nm cluster mode, 3-25 nm nucleation mode, 25-100 nm Aitken mode, and 100-1000 nm accumulation mode, as a function PM<sub>2.5</sub> concentration measured at BUCT-AHL in 2018 and 2019, for This figure starts 48 hours before CNY and runs through 48 hours after the CNY. The darker colorsfilled circles mark the nighttime measurements on the CNY (9pm-5am). The night-time mass concentrations are noticeably greater. The mass-to-number concentration comparison for CNY follows the same general curve during nighttime as the full time periodand the The pattern, including particularly the nighttime observations, is consistent with recent investigation by Zhou et al. ((2020), which showed that in general concentrations of pollutants are higher during nighttime, attributed to a lower boundary layer and consequent high concentrations within the boundary layer. As noted in Section 3.1, the 2020). The PM<sub>2.5</sub> concentrations during the CNY period in 2018 are significantly were nearly an order of magnitude higher than in other periods-before and after this time. The cluster and nucleation mode concentrations are lower. Aitken mode is on the same level, and accumulation mode clevated PM<sub>2.5</sub>

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concentration is higher during directly connected to the CNY period. This is reasonable, as elevated number concentration of accumulation mode particles contribute significantly to PM2.5 mass and constitute (Fig. 6 bottom right panel) and the majorCNY data points do not diverge from the overall coupling. This indicates that the typical sizes of particles contributing to PM2.5 remains similar during CNY than before and after it. Since the accumulation mode particle concentrations form the main part of the total particle surface acting as a condensation sink for vapors forming new particles in the atmosphere and a coagulation sink for small cluster and nucleation mode particles, it is natural that the concentrations of cluster and nucleation mode particles due to coagulation scavenging of smaller particles by larger ones, decrease with increasing PM2.5 (Fig. 6, upper panels).

In figure 5 we can see howshort, the CNY activities seem not to cause any major deviance for the typical aerosol dynamics other than the enhancement of the source of accumulation mode particles

Figure 7 depicts the cluster mode, nucleation, Aitken and accumulation mode particle number concentrations behaves as a function of sulfuric acid. Thegas phase sulfuric acid concentration in 2018 and in 2019 inside and outside of the CNY period. Looking at the clusters, the results show a general strong dependency on the sulfuric acid as it is one of the main pre-cursors driving the process of gas-to-particle conversion (e.g., Sipilä et al. 2010, Kulmala et al. 2013, Yao et al. 2018). However, the high nocturnal sulfuric acid concentration during CNY celebrations in 2018 dodoes 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 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.

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

Although fireworks Fireworks were formally prohibited within the 5th Ring Road of Beijing beginning in 2018, whereas outside the 5th Ring Road, there were no prohibitions (Liu et al., 2019). Still, there was stillsome evidence of fireworks firework burning either in the city or the immediate vicinity of the city, as measured atobserved BUCT-AHL, which is within the prohibition area. Nonetheless, because the initial peak during the 2018 CNY (Figure 6) 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.

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 MEP stations in the Beijing area from 2013 through 2019.

Figure 68 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, with the peak in PM<sub>2.5</sub> around midnight of the CNY reaching almost 700 μg/cm³ while values earlier in the day were less than 100 μg/cm³. The lowest levels of PM<sub>2.5</sub> were in 2019 with the overnight peak less than 200 μg/cm³ compared to daytime values around 50 μg/cm³. Observations from 2013, 2014, 2015, and 2017 also showed similarly high or higher levels of PM<sub>2.5</sub> 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 Years

day due to a network outage). The measurements for all seven years are in agreement with other studies that have linked elevated air pollution levels to CNY celebrations (Yang et al., 2014; Shi et al., 2014; Feng et al., 2012; Zhang et al., 2010), and this study further shows that the peak in 2019 is lower than in 2018, which is lower than in 2016 and 2017.

Data from the CNYs have also been compiled into box plots in Figure 79, depicting the distributions of pollutant concentrations from 6:00 pm on CNY Eve to 6:00 am on the CNY day each year, at all 12 MEP stations. The highest PM concentrations during this time were in 2016, and the 75th and 99th percentile concentrations have since decreased, after that. On the other hand, the median concentration remained high during 2017 and 2018 but decreased in 2019 by roughly a factor of two. Concentrations of NO2, and SO2, and CO all show a similar pattern as PM-more steady decrease than PM2.5, since the median concentration of both pollutants decreased steadily from 2016 (regarding NO2 for 2017), but for CO there is no clear pattern. It should be noted that in 2017, the data is missing after midnight, due to an unknown network outage. The decrease-more noticeable decrease in NO2 and SO2 is an expected outcome for a ban on firework burning, since both are produced by fireworks and have shorter lifetimes than CO and PM2.5 (Seinfeld and Pandis, 2016; Lee et al., 2011). Thus, they are less affected by long range transport and accumulation. The decrease in pollutant concentrations since 2016 agrees with the results obtained by Liu et al. (2019-). Since ozone is a secondary product and it reacts with several primary pollutants, its concentration pattern being roughly opposite to those of primary pollutants is as expected. In this aspect, it is notable that in 2019 in addition to primary pollutant concentrations also ozone concentration has decreased from 2017 and 2018.

Based on Hysplit back-trajectories (Figure 4), we see that in 2013-2015 the airmasses spent longer in the BTH area prior to arrival. This differs from the airmass sources in 2016-2017, where the airmasses come directly from the northwest. These areas to the northwest of Beijing, including Inner Mongolia and Mongolia usually contain less pollutants due to low anthropogenic emissions, and thus we can expect air masses from this region to be cleaner (Xu et al., 2008). Based on the airmass history, if emissions were the same, then there should be higher concentrations in 2013-2015; however, we see the highest concentrations of pollutants in 2016, followed by a decline after that. In 2018 and 2019, the airmasses spent around two days in the BTH area leading up to arrival at the station. Based on airmass source alone, we would have expected higher pollutant concentrations in 2018 and 2019, but this is not the case. Thus, we can conclude that emissions must have been highest in 2016, with lower emissions in 2018 and 2019. This agrees with Liu et al. (2019).

# 3.5 Spatial variability based on MEP measurement network data

A further analysis of the CNY in Beijing is to performNext, we performed a spatial comparison of the MEP measurements across the Beijing region. This includes comparing the observations inside the 5<sup>th</sup> Ring Road, where fireworks were prohibited, to outside the ring. Figure \$10 maps the 12 MEP stations in the Beijing region for 2013-2019, showing the relative difference of ratio between mean PM2.5 measurements concentration from 9 pm through 5 am during the night of CNY to and the average of measurements mean concentration within ± 48 hours of the CNY at each site. Figures \$2.813 in Supplementary Information show observations of PM2.5 from the 12 individual MEP sites and the corresponding differences, year-by-year from 2013-2019. Based on Figure \$10, we can see significant variation from year to year as to which station measures the highest pollution. It is important to note that the population density is greater closer to city center, and thus the population density could impact the results. However, it is plausible to assume that the relative population density difference between the city center and the surrounding areas do not change dramatically during the few yearyears' time period.

Figure <u>\$10</u> illustrates that in 2013 and 2014, the enhancement in PM<sub>2.5</sub> concentrations during CNY is greater inside the 5<sup>th</sup> ring than outside. In 2015, the enhancement is much greater at the two northeastern sites (HR and

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SY). In 2016, the differences vary, with no clear difference inside or outside the 5<sup>th</sup> ring. In 2018, the 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 had significantly high enhancement compared to the other sites. In 2019 the enhancement is overall less inside compared to outside. The enhancement factors outside the 5<sup>th</sup> Ring road (excluding the single highest value) and at the Northern inside stations nearest to the Ring road are quite similar in 2019, roughly in range 2.5 to 3, but the peak times of pollution are few hours earlier at the outside stations than the Northern inside stations (Fig. S12). The measurement sites closer to central Beijing, on the other hand, show clearly lower enhancement factors, of values of two or below. Based on these spatial and temporal differences, and on the northerly winds observed at the time, it is possible that the higher enhancement factors inside but close to the 5<sup>th</sup> Ring road are related to emissions from outside the Ring road.

Figure 911 shows differences between the PM<sub>2.5</sub> medianmean of the sites inside the 5th Ring Road and the medianmean of the sites outside the 5th Ring (that is the medianmean of the 8 inside sites minus the medianmean of the outside 4 stations) 48 hours before through 48 hours after the CNY for 2013-2019. In most years, there is a greater In 2013, 2014 and 2018, the enhancement of PM<sub>2.5</sub> during the CNY overnight is greater inside than outside, the 5th Ring Road. However, in 2015, the opposite was true. In and 2019, as well as immediately after the CNY midnight in 2016, the first part of the CNY overnight had PM2.5 was lower PM2.5 inside than outside, but that reversed a few hours later. In 2019, there was less. While we were lacking the detailed data on local meteorology during 2013-2016, we were still able to analyze the meteorological condition in terms of air mass trajectories. Figure 4 shows that, similar to 2019 as discussed previously, in 2015 and CNY midnight of 2016, airmasses arriving to Beijing were from the cleaner West-North sector and arrived with much higher velocity in comparison to years 2013, 2014 and 2018, during which the air masses made a turn in South or East before arrival to Beijing. Even though the CNYs during which the increase of PM2.5 enhancement inside the 5th Ring Road is less pronounced than outside throughout the seem to be related to faster arrival of cleaner airmasses, we have no clear view for the reason of this difference and, due to the qualitative nature of this comparison, it is well possible that this connection is pure coincidence. The similarity of years 2015 and 2019 in terms of the spatial variation of CNY midnight pollution peak suggests that meteorology may be at least part of the reason for the lesser enhancement of pollution levels inside the 5th Ring Road than outside. Nevertheless, the notably lower concentrations of PM2.5 and gaseous air pollutants in 2019 than in 2015 indicates that, even with similarities in spatial distribution of changes in concentrations, the most likely reason for lower concentrations during CNY night of CNY. are the lower emissions.

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#### 4 CONCLUSIONS

ThisIn this study, we looked at comprehensive measurements over CNY 2018 and 2019 at a measurement station in Beijing, along with long-term datasets across the Beijing metropolitan area.

Our study confirms that CNY consistently impacts air quality in Beijing year after year. These, Based on our observations at BUCT-AHL station in Beijing, in 2018, we detected higher than typical night-time concentrations of particulate mass (PM<sub>2.5</sub>), particle number, trace gas and sulfuric acid concentrations during the CNY. This was expected, and these results are consistent with previous studies that have linked the CNY (and other similar holiday celebrations involving firework burning around the world) to degraded air quality both locally and regionally. Our results suggest that the regulations to limit firework use have improved the air quality within the restriction zone inside the fifth ring road in Beijing since 2016.

Based on our observations at BUCT AHL station Our results suggest that the regulations from CNY 2018 to limit firework use have improved the air quality within the restriction zone inside the 5<sup>th</sup> Ring road in Beijing, and from 2016 to 2019 there has been a decrease in the effects of holiday-related pollution, which offers an

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optimistic outlook to the air quality impacts caused by CNY and the consequential public health concerns stemming from air pollution.

During the CNY night in 2018, we detected clearly higher than typical night time concentrations of particulate mass (PM2.5), particle number, trace gas and sulfuric acid concentrations during the CNY. The increase in sulfuric acid concentration did not lead to observed new particle formation, which is explained by simultaneously increasing condensation and coagulation sinks for clustering vapors and freshly formed particles, respectively. However, we observed appearance of particles with diameters of roughly 20100 nm that seemed to be linked to enhanced sulfur dioxide, sulfuric acid and black carbon concentrations, most likely as a result from firework burning. Based on the MEP data, the peaks in concentrations of different pollutants were noticeably lower than in the previous years. In 2019, a peak in pollution was observed overnight, but it was significantly lower than in 2018, while meteorological conditions were comparable in both years. —The significant year-to-year variability depended presumably on the meteorological conditions, on new imposed regulations as well as on the fact that the CNY period is determined with a lunar calendar and therefore the exact CNY period varies from year to year. In 2013, 2014, and 2015, haze episodes lasting several days were observed immediately following the CNY. In 2016 and . A common phenomenon for both 2018 and 2019 CNY nights was the accumulation of secondary aerosol throughout the night, seen as a diameter growth of the dominant particle mode in particle number size distributions. Measurements at BUCT-AHL showed that in 2018 a moderate haze episode began one day following the CNY, potentially related to the firework burning.

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

ThisTo conclude, this long-term analysis, which combines BUCT data with multiple years of Chinese government data at 12 locations in the Beijing area, demonstrates the importance of analyzing multiple data sources to determine overall trends, rather than making conclusions based on a single dataset. This also demonstrates the usefulness of long-term measurements. Therefore, we suggest ongoing measurements at both BUCT AHL and MEP sites into multiple future years.

The combination of the BUCT AHL comprehensive observations together with the spatial variability provided by the MEP sites Using these datasets together, we see excellent potential that can be utilized to investigate the changes in a) atmospheric chemistry – likesuch as 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 Using CNY as a case study offers excellent insight into how rapid changes in emissions will affect air quality, health, and quality of life, especially in megacities such as Beijing. To confirm and quantify the influence of banning the firework burning in Beijing and the impact of varying meteorological conditions, similar data from coming CNYs is needed. Therefore, we suggest ongoing measurements at both BUCT-AHL and MEP sites into multiple future years.

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**Author Contributions** 

 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, PP 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 with assistance from SG. BF wrote the original draft and prepared the manuscript. PP, TP and all other authors reviewed and edited the manuscript.

**Competing Interests** 

The authors declare that there are no conflicts of interest in this study.

**Data Availability** 

Data from the BUCT station is available at request. Real time data from the MEP stations is available at http://106.37.208.233:20035/. Archived, quality-controlled MEP data may also be available upon request.

REFERENCES

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.

Barman, S. C., Singh, R., Negi, M. P. S., and Bhargava, S. K. (2007). Ambient air quality of Lucknow City (India) during use of fireworks on Diwali Festival. *Environ. Monit. Assess.*, 137:495–504.

Chen, B., Kan, H., Chen, R., Jiang, S., and Hong, C. (2011). Air Pollution and Health Studies in China—Policy Implications. *Journal of the Air & Waste Management Association*.65-11:1292-1299.

Chen, Z., Zhang, J., Zhang, T., Liu, W., and Liu, J. (2015). "Haze observations by simultaneous lidar and WPS in Beijing before and during APEC, 2014." *Science China Chemistry*. 58:1385–1392.

Chou, C. C.-K., Tsai, C.-Y., Shiu, C.-J., Liu, S. C., and Zhu, T. (2009). Measurement of NO<sub>y</sub> during Campaign of Air Quality Research in Beijing 2006 (CAREBeijing-2006): Implications for the ozone production efficiency of NO<sub>x</sub>. *Journal of Geophysical Research: Atmospheres.* 114:D00G01.

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Dada, L., Ylivinkka, I., Baalbaki, R., Li, C., Guo, Y., Yan, C., Yao, L., Sarnela, N., Jokinen, T., Daellenbach,
K. R., Yin, R., Deng, C., Chu, B., Nieminen, T., Wang, Y., Lin, Z., Thakur, R. C., Kontkanen, J., Stolzenburg,
D., Sipilä, M., Hussein, T., Paasonen, P., Bianchi, F., Salma, I., Weidinger, T., Pikridas, M., Sciare, J., Jiang,
J., Liu, Y., Petäjä, T., Kerminen, V.-M., and Kulmala, M. (2020). "Sources and sinks driving sulfuric acid
concentrations in contrasting environments: implications on proxy calculations." Atmos. Chemistry and
Physics, 20:11747–11766.

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> during the 2009 Chinese new year's holiday in Shanghai. *Atmospheric Research*, 118:435–444.

Guo, B., Wang, Y., Zhang, X., Che, H., Zhong, J., Chu, Y., and Cheng, L. (2020). "Temporal and spatial variations of haze and fog and the characteristics of PM2.5 during heavy pollution episodes in China from 2013 to 2018." *Atmospheric Pollution Research*. 10:1847-1856

Hari, P. and Kulmala, M. (2005). Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II). *Boreal Environment Research*. 10:315-322.

He, H., Li, C., Loughner, C. P., Li, Z., Krotkov, N. A., Yang, K., Wang, W., Zheng, Y., -Bao, X., Zhao, G., and Dickerson, R. R. (2012). SO<sub>2</sub> over central China: Measurements, numerical simulations and the tropospheric sulfur budget. *Journal of Geophysical Research*, 117, D00K37.

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.

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.

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 air quality: insights of tracers, source evolution and aging processes. *Atmospheric Chemistry and* 

708 air quality: insights of 709 *Physics*.15:2167-2184.

Kulmala, M., Kontkanen, J., Junninen, H., Lehtipalo, K., Manninen, H.E., Nieminen, T., Petäjä, T., Sipilä, M., Schobesberger, S., Rantala, P., Franchin, A., Jokinen, T., Järvinen, E., Äijälä, M., Kangasluoma, J., Hakala, J., Aalto, P.P., Paasonen, P., Mikkilä, J., Vanhanen, J., Aalto, J., Hakola, H., Makkonen, U., Ruuskanen, T.M., Mauldin III, R.L., Duplissy, J., Vehkamäki, H., Bäck, J., Kortelainen, A., Riipinen, I., Kurtén, T., Johnston, M.V., Smith, J.N., Ehn, M., Mentel, T.F., Lehtinen, K.E.J., Laaksonen, A., Kerminen, V.-M. and Worsnop, D.R. (2013). Direct observations of atmospheric nucleation, *Science*, 339, 943-946- (2015). Atmospheric Chemistry: China's Checking Cocktail. *Nature Comment*,

Kulmala, M. (2015). Atmospheric Chemistry: China's Chocking Cocktail. Nature.

Kulmala, M., Kerminen, V.-M., Petäjä, T., Ding, A.J. and Wang, L. (2017). Atmospheric Gas-to-Particle Conversion: why NPF events are observed in megacities? *Faraday Discussions*, 200, 271-288, doi: 10.1039/c6fd00257a.

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725 Kulmala, M. (2018). Build a global Earth observatory. Nature-Comment.

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*. 116:6375-

729 6386.

Lee, C., Martin, R. V., van Donkelaar, A., Lee, H., Dickerson, R. R., Hains, J. C., Krotkov, N., Richter, A.,
 Vinnikov, K., and Schwab, J. J. (2011), SO<sub>2</sub> emissions and lifetimes: Estimates from inverse modeling using
 in situ and global, space-based (SCIAMACHY and OMI) observations, J. Geophys. Res., 116, D06304,
 doi:10.1029/2010JD014758.

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.

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.

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 and Environmental Safety*. 171:406-413.

Liu, Y.C., Yan, C., Feng, Z., Zheng, F., Fan, X., Zhang, Y., Li, C., Zhou, Y., Lin, Z., Guo, Y., Zhang, Y., Ma, 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 environment." Big Earth Data 4, 295-321.

Manninen, H. E., Mirme, S., Mirme, A., Petäjä, T., and Kulmala, M. (2016). How to reliably detect molecular clusters and nucleation mode particles with Neutral cluster and Air Ion Spectrometer (NAIS), *Journal of Atmospheric Measurement Techniques*, 9:3577-3605, 10.5194/amt 9-3577-2016, 2016.

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.

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.

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

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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.

Seinfeld, J. and Pandis, S. (2016). Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 3rd Edition. ISBN: 978-1-118-94740-1

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.

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.

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.

Sipilä, M., Berndt, T., Petäjä, T., Brus, D., Vanhanen, J., Stratmann, F., Patokoski, J., Mauldin III, R.L., Hyvärinen, A.-P., Lihavainen, H. and Kulmala, M. (2010). The Role of sulfuric acid in atmospheric nucleation, *Science*, 327, pp. 1243-1246.

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.

Stein, A. F.; Draxler, R. R.; Rolph, G. D.; Stunder, B. J. B.; Cohen, M. D.; Ngan, F. (2015). "NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System". *Bulletin of the American Meteorological Society*. 96 (12): 2059–2077.

Sun, Y., Wang, Z., Fu, P., Jiang, Q., Yang, T., Li, J., and Ge, X. (2013). "The impact of relative humidity on aerosol composition and evolution processes during wintertime in Beijing, China." *Atmospheric Environment*. 77: 927-934.

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.

Vanhanen, J., Mikkilä, J., Lehtipalo, K., Sipilä, M., Manninen, H. E., Siivola, E., Petäjä, T., and Kulmala, M. 817 (2011). Particle Size Magnifier for Nano-CN Detection. *Aerosol Science and Technology*, 45:533-542,

818 10.1080/02786826.2010.547889.

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Virkkula, A., Chi1, X., Ding, A., Shen, Y., Nie, W., Qi, X., Zheng, L., Huang, X., Xie, Y., Wang, J., Petäjä,
 T., and Kulmala, M. (2015). On the interpretation of the loading correction of the aethalometer. <u>Atmospheric</u>
 Measurement Techniques. 8:4415–4427.

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Wang, F., D. S. Chen, D.S., Cheng, S.Y., Li, J. B., Li. M. J., and Ren, Z. H. (2010). "Identification of regional atmospheric PM<sub>10</sub> transport pathways using HYSPLIT, MM5-CMAQ and synoptic pressure pattern analysis."
 Environmental Modelling & Software. 25,8:927-934.

Wang, Y., Zhuang, G., Xu, C., and Ana, Z. (2007). "The air pollution caused by the burning of fireworks during the lantern festival in Beijing." *Atmospheric Environment*. 41-2:417-431.

Weather Underground. "Beijing, People's Republic of China Weather History" https://www.wunderground.com/history/weekly/cn/beijing/ZBNY/date/2019-2-4

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.

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.

Xu, X., Barsha, N. and Li, J. (2008). "Analyzing Regional Inluence of Particulate Matter on the City of Beijing, China." *Aerosol and Air Quality Research*. 8:78-93.

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 Chinese New Year celebration in Jinan, China. *Science of the Total Environment*, 476-477:57–64.

Yao, L., Garmash, O., Bianchi, F., Zheng, J., Yan, C., Kontkanen, J., Junninen, H., Mazon, S.B., Ehn, M., Paasonen, P., Sipilä, M., Wang, M.Y., Wang, X.K., Xiao, S., Chen, H.F., Lu, Y.Q., Zhang, B.W., Wang, D.F., Fu, Q.Y., Geng, F.H., Li, L., Wang, H.L., Qiao, L.P., Yang, X., Chen, J.M., Kerminen, V.-M., Petäjä, T., Worsnop, D.R., Kulmala, M. and Wang, L. (2018). Atmospheric new particle formation from sulfuric acid and amines in a Chinese megacity. *Science*, 361, 278-281.

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 site, Hyderabad, India, during three consecutive years. *Environ. Monit. Assess.*, 185:7309–7325.

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 Environment*, 44:5191–5198.

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.

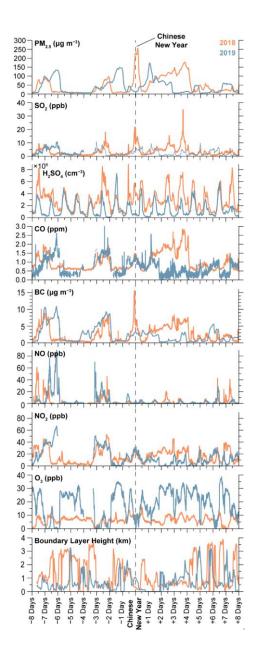
Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W. W., Dong, F., He, D., Shi, Q. F. (2013). Analysis of a winter
 regional haze event and its formation mechanism in the North China Plain. *Atmospheric Chemistry and Physics*.
 13:5685-5696.

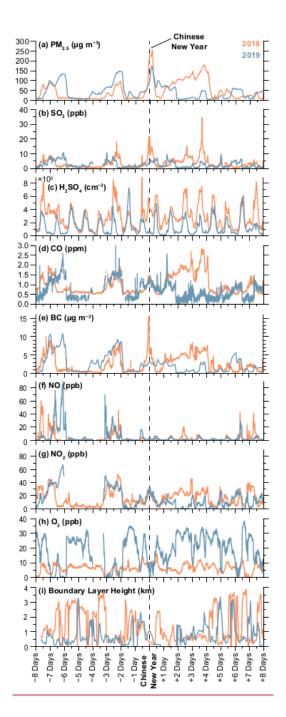
Zhou, Y., Dada, L., Liu, Y., Fu, Y., Kangasluoma, J., Chan, T., Yan, C., Chu, B., Daellenbach, K. R., Bianchi,
F., Kokkonen, T. V., Liu, Y., Kujansuu, J., Kerminen, V.-M., Petäjä, T., Wang, L., Jiang, J., and Kulmala, M.
(2020). Variation of size-segregated particle number concentrations in wintertime Beijing. *Atmospheric Chemistry and Physics*. 20:1201–1216

Zhu Y. Y., Gao Y. X., Chai W. X., Wang, S., Li, L., Wang, W., Wang, G., Liu, B., Wang, X. Y., Li, J. J. (2020). "Heavy Pollution Characteristics and Assessment of PM2.5 Predicted Model Results in Beijing-Tianjin-Hebei Region and Surrounding Areas During November 23 to December 4, 2018". DOI: 10.13227/j.hjkx.201908123.



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





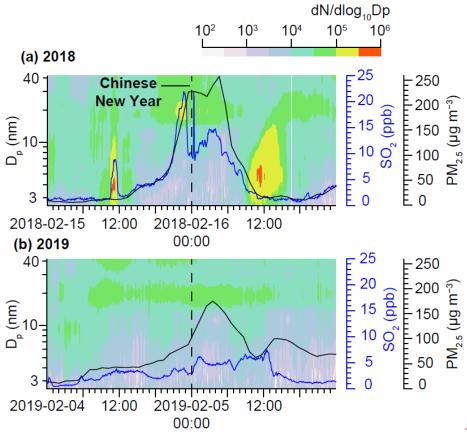


Figure 3: Aerosol number size distribution from NAIS instrument

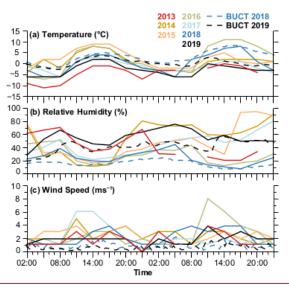
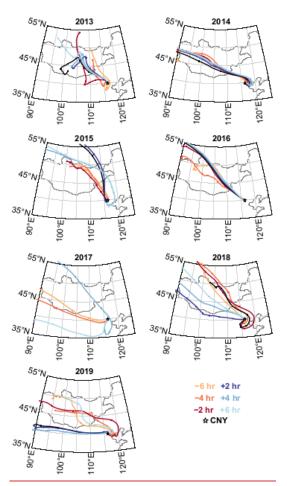


Figure 3: Meteorological conditions during CNY night ± one day measured in Beijing from 2013-2019. Solid lines are measurements from Beijing Nanyuan Airport (ZBNY). These measurements are every three hours. Dashed lines are measurements at BUCT-AHL during 2018-2019, with time resolution of one hour.



**Figure 4:** Hysplit 96 hour back-trajectories for airmasses arriving at BUCT-AHL between 18:00 and 06:00 local time, the night of CNY in 2013 through 2019. The markers are every 12 hours.

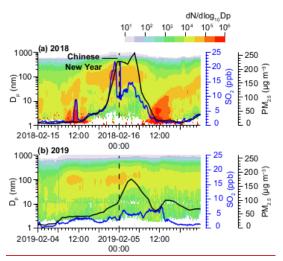
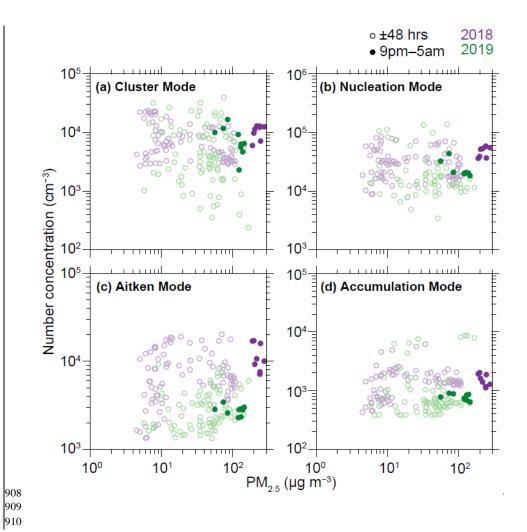


Figure 5: Aerosol particle number size distribution (PSD) from one day before the CNY through one day following the CNY in 2018 and 2019, overlain with aerosol mass concentration PM<sub>2.5</sub>, black lines) and SO<sub>2</sub> (blue lines). A spike of PM<sub>2.5</sub> and SO<sub>2</sub> is observed 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. (black lines) and SO<sub>2</sub> (blue lines).



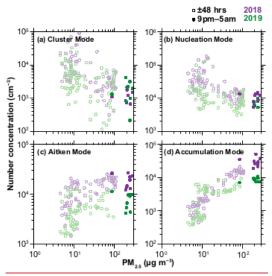
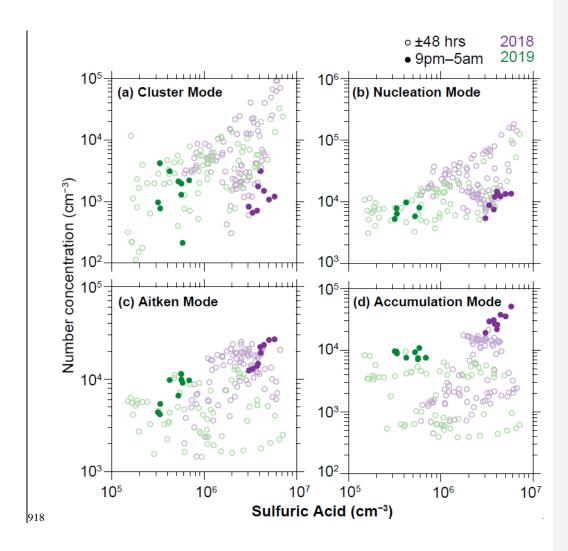


Figure 4: PM<sub>2.5</sub>6: Aerosol particle number econcentration as a function of mass concentration concentrations in cluster, nucleation, Aitken<sub>7</sub> and accumulation modes in—as a function of PM2.5 mass concentration in 2018 (purple) and 2019, with comparison of CNY  $\pm$  48 hours with measurements (green), separated from 9pm through 5am the night of the CNY. Measurements are (filled circles) and those of CNY  $\pm$  48 hours (open circles). The data is from BUCT-AHL.



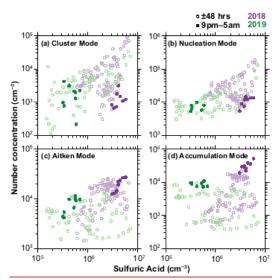
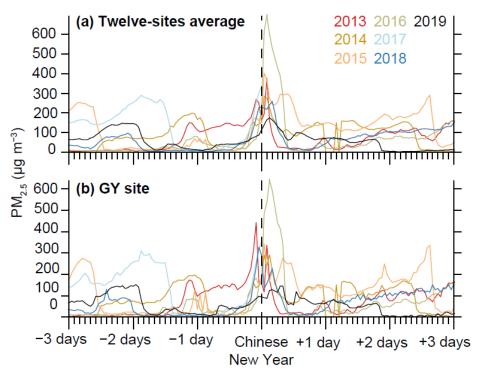


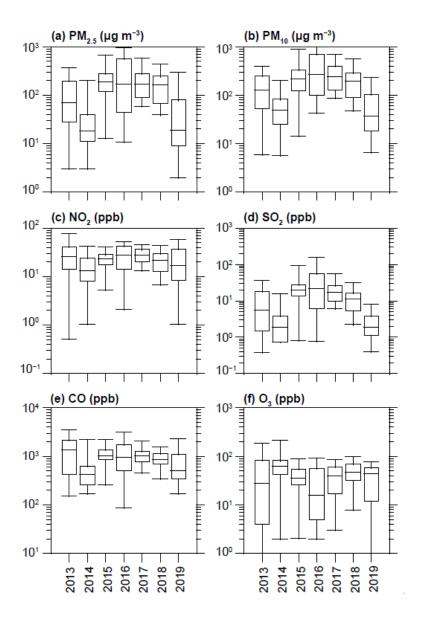
Figure 5: Number concentration of particles7: Aerosol particle number concentrations in cluster, nucleation, Aitken, and accumulation modes as a function of gas phase sulfuric acid concentration in 2018 (purple) and 2019, with comparison of CNY  $\pm$  48 hours with measurements (green), separated from 9pm through 5am the night of the CNY. Measurements are (filled circles) and those of CNY  $\pm$  48 hours (open circles). The data is from BUCT-AHL.

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**Figure 68:** PM<sub>2.5</sub> 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.

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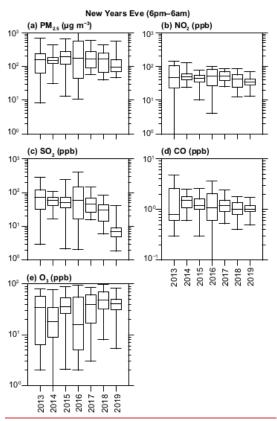
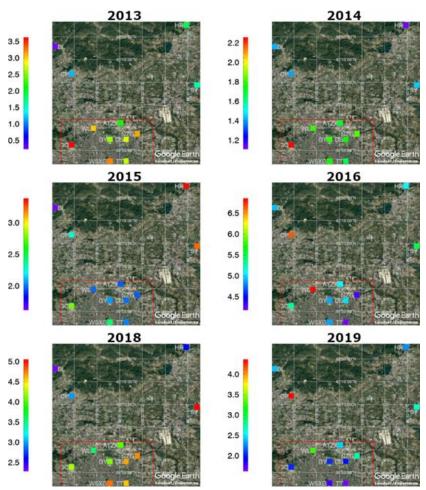
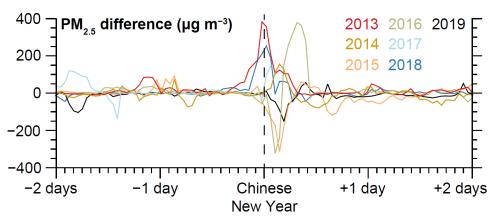


Figure 79: Boxplots of particulate matter  $PM_{2.5}$  and trace gases between 18:00 and 06:00 on the night of the Chinese New Year in the years 2013-2019. The boxplots show  $1^{st}$ ,  $25^{th}$ ,  $50^{th}$ ,  $75^{th}$ , and  $99^{th}$  percentiles of the data across the 12 sites during this 12-hour period (13 time points, inclusively).



**Figure \$10**: 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.



**Figure 911:** Differences <u>ofbetween mean PM<sub>2.5</sub> concentrations</u> inside <u>vs.and</u> outside the 5<sup>th</sup> Ring Road of Beijing from 2013 through 2019. <u>Positive values indicate higher concentration inside 5<sup>th</sup> Ring Road.</u>