Response to reviewer comments:

General response:

We thank both reviewers for providing constructive comments.

We have carefully considered every single comment and revised the manuscript accordingly. We also provided a point by point response to all the comments made below.

One point we would like to make is that this is an Introduction to special issue paper, not an overview or research paper. ACP editorial policy states that "*Special issues may include an introduction article or an overview article or both. Introduction articles* ***outline the motivation and background****, and overview articles synthesize and summarize the findings of the special issue papers. The manuscript title must clearly reflect the relation to the special issue and should start with "Introduction:" or "Overview*".

To make this clearer, we have added a paragraph at the end of the introduction.

"This introduction paper describes the motivation and background of APHH-Beijing programme, and presents some of the background air quality and meteorology observations that lay the basis of data interpretation for the whole programme, particularly during the two intensive field campaigns. These campaigns form one of the core research activities within APHH-Beijing integrating the different themes / projects. We did not intend to present the key scientific results of APHH-Beijing here as much of the research activities are still ongoing and unpublished. Such information is more suitable to go to an overview paper."

We also would like to emphasize that scientific work on the impact of synoptic scale meteorology on air quality and the air quality climatology add significant knowledge to our understanding of air pollution events in Beijing. Therefore, this introduction paper not only provides the motivation and background of the APHH-Beijing programme but also new science.

Many of the ACP special issues have introduction papers, such as:

Kulmala, M. et al., 2009. Introduction: European Integrated Project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales. Atmos. Chem. Phys., 9, 2825–2841.
Cairo, F., et al., 2010. An introduction to the SCOUT-AMMA stratospheric aircraft, balloons and sondes campaign inWest Africa, August 2006: rationale and roadmap. Atmos. Chem. Phys., 10, 2237–2256
Kruger, K and Quack, B., 2013. Introduction to special issue: the TransBrom Sonne expedition in the tropical West Pacific. Atmos. Chem. Phys., 13, 9439–9446
Kulmala, M. et al., 2015. Introduction: The Pan-Eurasian Experiment (PEEX) – multidisciplinary, multiscale and multicomponent research and capacity-building initiative. Atmos. Chem. Phys., 15, 13085–13096
Martin, S.T. et al., 2016. Introduction: Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5). Atmos. Chem. Phys., 16, 4785–4797.

Reviewer 1

Comment 1: several giant projects on air pollution and health impacts funded in the volume of billions RMB yuan are processing currently or have been completed in Beijing and neighboring provinces in the last decade. These works should be summarized to make the literature review more complete. The authors are strongly encouraged to present a summary to highlight the importance of APHH-Beijing in comparison with others.

Response: We agree that a summary of past work in Beijing will be valuable to put the APHH-Beijing work into context and we have added a summary about the CARE-Beijing and other large programmes (see below). APHH-Beijing programme was designed in 2015 and started in 2016. The rationale of this programme in the introduction paper was based on work up to 2016. Thus, we feel that it is not totally appropriate to include ongoing and unpublished work in this introduction paper.

Changes in the texts: We have added texts in the Introduction

"Many research programmes were initiated in Beijing to study the air pollution processes since late 1990s. Earlier research programmes (e.g., early 2000) focused on primary emissions of SO2, NO2, CO, PM10, volatile organic compounds and then secondary pollutants such as ground-level ozone and secondary fine particles. These researches contributed to the development of air pollution mitigation strategies by the Beijing Municipal government.

Beijing Olympic Games (2008) offered additional incentives to improve air quality and this led to the funding of CAREBEIJING (Campaigns of Air Pollution Research in Megacity Beijing and Surrounding Region) and other major programmes. CAREBEIJING was initiated and organized by Professor Tong Zhu of Peking University, with participation of hundreds of scientist and students from China, USA, Germany, Italy, Japan, and South Korea. The field campaigns were conducted in the summer of 2006, 2007, and 2008, with the objectives to learn the environmental conditions of the region, to identify the processes (transport and transformation) that lead to the impact of the surrounding area on air quality in Beijing, to quantify the impact of the surrounding area on air quality in Beijing, and to formulate policy suggestion for the air quality attainment during the 2008 Beijing Olympic Games. Other major research programmes, initiated since early 2000, aimed to provide scientific basis to deliver air pollution mitigation measures for ensuring a good air quality during the Olympics Games. Measures developed as a result of these programmes successfully reduced the air pollution during the Olympics Games, and provided valuable examples for air pollution control policy-making in other cities (Wang et al., 2010). CARE-BEIJING latter on was extended to CAREBEIJING-NCP (Campaigns of Air Pollution Research in Megacity Beijing and North China Plain), where field campaigns were carried out in the summer of 2013 and 2014 to investigate the transport and transformation processes of air pollutants in megacity Beijing and North China Plain. The results of CAREBEIJING and CAREBEIJIN-NCP have been published in three special issues of Atmospheric Chemistry and Physics (https://www.atmos-chem-phys.net/special_issue198.html) and Journal of Geophysical Research-Atmospheres (https://agupubs.onlinelibrary.wiley.com/doi/toc/10.1002/(ISSN)2169-8996.CARBS1). These large research programmes and numerous discovery science projects significantly enhanced our understanding on the emission, sources and processes of air pollutants in Beijing (Chan and Yao, 2008; Zhu et al., 2012). However, our understanding of sources and emissions of key air pollutants such as PM2.5 and ozone and the role of the interactions between physical and chemical processes in the formation of pollution events in the Beijing megacities is still far from being accurate or complete. In addition, none of the abovementioned large programmes are directly linked heath effect studies. "

In addition, we have added a section at the end of the paper to summerize this introduction paper and highlight the novel aspects of the APHH-Beijing.

"The APHH-Beijing is an integrated and multidisciplinary research programme by leading UK and China researchers to (1) quantify sources and emissions of urban atmospheric pollutants; (2) elucidate processes affecting urban atmospheric pollution events; (3) estimate the personal exposure and impacts of air pollution on human health, and (4) develop intervention strategies to improve air quality and reduce health impacts in the Beijing megacity. This introduction paper outlines the motivation of the APHH-Beijing programme as well as provides the background air quality and meteorological conditions that form the basis of data interpretation for the whole programme, particularly during the two intensive field campaigns as a core research activity within the programme.

APHH-Beijing has measured the fluxes of key air pollutants, including NOx, CO, BC, VOCs and speciated particulate matter, applied a suite of traditional and modern techniques to apportion the sources of particulate matter, determined a wide range of pulmonary and cardiovascular biomarkers linking to direct personal exposure and extensive fixed-station monitoring as well as source apportionment results, and evaluated the effectiveness of Beijing's air pollution control policies using both chemical transport models and novel machine learning techniques. A number of papers have already been published under the APHH-China programme including those in APHH-Beijing special issue (Wang et al., 2019; Pan et al., 2019; Xia et al., 2018; Zhou et al., 2018; Wang et al., 2018; Lyu et al., 2019; Hollaway et al., 2019; Du et al., 2018; Liu et al., 2018a,b; Smith et al., 2018). More papers are being prepared for publication in this special issue and elsewhere, which will cover (but not limited to) emission fluxes of air pollutants, chemical composition and source apportionment of fine particles, satellite observations of trace gases and aerosols, sources and processes leading to haze events and photochemical smogs, physical and optical properties of aerosol particles, formation processes of secondary aerosols, urban meteorology, feedbacks between haze, photochemistry and meteorology, integrated regional and urban scale modelling, personal exposure to air pollutants and human health effects of air pollution."

Comment 2: Section 2 is too ambitious to be practical for two short-term campaigns

Response: It appears that there is a misunderstanding here. The whole programme is more than the two campaigns. We have introduced the two campaigns because they are one of the core activities that integrate research across the themes and the information provided in this introduction paper provided a background for a number of in-preparation papers for this special issue.

Section 2 is extracted from the five funded proposals that were awarded on a competitive basis and assessed by international expert reviewers and a panel of UK/China top scientists. Now look back at these set objectives, we have indeed make progresses in all areas.

Comment 3: Lines76-80 "The winter campaign was characterized by high PM2.5 pollution events whereas the summer experienced high ozone pollution events. Air quality was poor during the winter campaign, but less severe than in the same period in 2015 when there were a number of major pollution episodes. PM2.5 levels were relatively low during the summer period, matching the cleanest periods over the previous five years." The statement looks like the report issued by local EPD rather than a scientific study. The reviewer gains almost nothing from it. It should be more specific.

Response: This is a very general introduction which sets our campaign periods in context. However, we recognise that it is very qualitative and have therefore modified it to include quantitative information which guides the reader more usefully. The revised text reads as follows:

Changes made:

"The winter campaign was characterised by high PM2.5 pollution events with peak hourly concentrations at the urban site ranging up to 498 µg m^{-3}, whereas the summer experienced events of high ozone concentrations with the highest hourly average up to 176 ppb. Air quality was generally poor during the winter campaign with an average PM2.5 concentration of 96 µg m^{-3}, but less severe than in the same period in 2015. Synoptic scale meteorological analysis suggests that the greater stagnation and weak southerly circulation in November/December 2016 contributed to the poor air quality during all haze events detected. PM2.5 levels were relatively low during the summer campaign with the highest daily concentration of only 79 µg m^{-3}, matching the cleanest periods over the previous five years."

Comment 4: Lines 80-82, "Synoptic scale meteorological analysis suggests that the greater stagnation and weak southerly circulation in November/December 2016 may have contributed to the poor air quality." Contributed to a few or all severe PM2.5 pollution events?

Response: We updated Figure 12 to include indication of haze events. This clearly demonstrates that CTs associated with stagnation (CT9, 11) dominate during all haze events of the winter campaign. The text and abstract have been updated accordingly.

The sentence in the abstract is now changed to
"*Synoptic scale meteorological analysis suggests that the greater stagnation and weak southerly circulation in November/December 2016 ~~may have~~ contributed to the poor air quality during all haze events detected."*

Comment 5: Line 100, "particularly severe in developing megacities, such as Beijing, where rapid urbanisation has led to a fast increase in pollution emissions (Guan et al., 2014), on top of regional pollution from industrial and other anthropogenic activities." Can Beijing be called as developing megacities? The reviewer also cannot understand the statement, please consider to revise.

Response*:* We recognize that the definition of a "developing megacity" can sometimes be controversial. In this context, we argue that Beijing is a developing megacity because it is still transforming rapidly and its GDP growth is significantly faster than developed megacities. We have revised the sentence:

Changes made: The quoted sentence has now been changed to:
"*Air pollution is particularly severe in developing megacities, such as Beijing, where pollutants from traditional sources, such as solid fuel combustion are mixed with those from road traffic (Guan et al., 2014), on top of regional pollution from industrial and other anthropogenic activities."*

Comment 6*:* Lines 117-119 "This makes Beijing a particularly interesting place to study as it provides a new environment to test our understanding of urban pollution processes." The reviewer feels very surprised that all Chinese co-authors agree with the statement.

Response: We have revised this sentence as following:
"*This makes Beijing a particularly interesting place to study as it provides an atmospheric environment very different to developed megacities such as London and Paris to investigate urban pollution processes."*

Comment 7: The objectives in Section 2 are ambitious. The reviewer has doubt how they can be achieved through two short-term campaigns at two sites.

Response: see above response to comment 2.

Comment 8: Section. 3.1.1 does not sound scientific to this reviewer. It makes more sense to use the data from the air quality monitoring network and the two additional sites together to evaluate the accuracy of emissions of air pollutants?
Response: The air quality monitoring network is a valuable source of data but measures only a small suite of classical pollutants whereas our monitoring campaigns measured a much larger range of species, which are helpful in constraining the numerical models. We also made air pollutant flux measurements at the IAP site and that can only be done with a tower. We have also been analysing data from the monitoring network in Beijing and agree that this is a valuable resource in model validation studies. Consequently, we have modified the final paragraph of Section 3.1.1 to read as follows:

"Measured ground level concentrations both from our campaign sites and the Beijing monitoring network, together with source apportionment results, are compared with the predictions of a chemistry-transport model and used to provide a clear distinction between advected regional pollution and the impact of local sources…".

Comment 9: Lines 243-246" Previous studies of pollution in Beijing have shown that it is often perturbation of the physicochemical and dynamic atmospheric conditions that modulate the most severe air quality events, rather than changes in emissions, for example during the development of stable inversions or periods of strong photochemistry." The references are missing. Please consider to revise. The statement is hard to follow

Response: we revised this sentence to :

"Previous studies of pollution in Beijing have shown that the interactions of physical conditions, such as the development of temperature inversion in the atmosphere, and chemical processes, e.g., formation of secondary pollutants, such as aerosol particles and ozone that modulate the most severe air quality events."

Comment: Lines 280-283, " AIRLESS aimed to advance air quality and health research in China by bringing together two fields of research that have made rapid advancements in recent years: measurements of a wide range of pulmonary and cardiovascular biomarkers in a panel study and personal monitoring of multiple air pollutants with high spatio-temporal resolution by sensor technology" In China or In Beijing and Neighboring Provinces? Why are the two sites' measurements helpful for the targets?
Response: All work is done at Beijing. We recognize that we could have been more specific. We have revised this sentence as below. We have identified the reasons why the site measurements are useful for AIRLESS (see below).

" AIRLESS aimed to advance air quality and health research in Beijing by bringing together two fields of research that have made rapid advancements in recent years: measurements of a wide range of pulmonary and cardiovascular biomarkers in a panel study and personal monitoring of multiple air pollutants with high spatio-temporal resolution by sensor technology"

"AIRLESS is also benefiting from the use of an extensive range of pollution metrics and source apportionment results collected from in the Themes 1 and 2 projects. "

Reviewer 2:

Comment 1: However, to be qualified as an ACP research article, the authors need to provide more sciences in the manuscript, as suggested below.

Response: This is not a research article but an introduction paper. Please see the general response above.

Comment 2: There have been many field campaigns, e.g., CAREBeijing, organized in the past 10 years targeted on investigating the air pollution and its health impact in Beijing. Compared with all those previous studies, what is unique about the current project? What are the scientific challenges this project aims to solve?
Response: Please see response to comment 1 by reviewer 1. APHH-Beijing is unique in that it is an integrated programme quantifying emissions of air pollutants using bottom-up, tower-based flux and satellite measurements, apportioning sources of particulate matter by multiple receptor models and hybrid receptor-chemical transport models, understanding atmospheric processes leading to pollution events via a coordinated measurements of physics and chemistry, and quantifying the health effects to individuals through personal exposure and health indicator / novel metabolomics measurements. APHH-Beijing integrates strengths in atmospheric sciences the UK with emerging research capabilities in China. In the newly added summary, we also highlighted the novel aspects of APHH-Beijing programme. Please also see response to reviewer comment 1.

Comment 3: There are four research themes presented: sources and emissions, atmospheric processes, health effects, and solutions. The last two only appeared in the very first part of the manuscript, no scientific output can be found later on. To make this manuscript completer and more consistent, primary results related to the health impacts need to be given.

Response: This is an introductory paper designed to set the scene rather than to give the conclusions of the study. Consequently, we feel that it would be inappropriate to report results from the health impacts studies which have yet to be subjected to peer review of their findings. It is our intention to write an overview paper towards the end of the APHH-Beijing programme where we will summerize the outcomes of the whole programme.

Comment 4: One focus of this manuscript is the overview of two joint field campaigns. Indeed, there are lots of discussions regarding the site information and type of instruments, but these discussions are not necessarily useful, as any future publications related to these two campaigns would have to give similar descriptions in their methods section anyway. Instead, this manuscript could be a nice platform for a detailed instrument calibration and comparison, data analysis and uncertainty quantification, and so on.

Response: One of the reasons why we have provided detailed site information is exactly to avoid every single paper to have a long paragraph describing exactly the same information. All further papers can refer to this paper for site information.

The list of instruments are very important part of the introduction paper as this gives readers an overview (big picture) of what is being measured within APHH-Beijing and see immediately if they might be able to find a particular type of data that they are interested in. Most single projects will make some particular type of measurements, but the APHH-Beijing programme made many complementary measurements.

We feel that instrument calibration is a routine work by each research group and this is too detailed for an introduction paper. Instead, such information should go to individual papers.

We take intercomparison extremely seriously within APHH-Beijing, particularly for those species that are still hard to measure, such as HONO. It is our intention that such highly specialized subjects will be published in individual papers rather than in this introduction paper. For example, an intercomparison HONO dataset has been generated for the whole APHH-Beijing programme to use, including both modelling and measurement scientists.

Data analysis and uncertainty quantification are highly complex in particularly when we are considering hundreds of species are being made. It is impractical to include such detailed information within an introduction paper.

Comment 5: The last two sections describe the air quality, e.g., the average concentrations and diurnal patterns of common air pollutants like NOx, O3, PM2.5, and etc., during the two field campaigns. As the authors highlighted earlier that regional modeling is an essential part of the campaigns, a modeling vs. observation comparison in terms of temporal profiles of these common pollutants need to be provided.
Response: We highly value this constructive comment and have added section 7 to address this comment. We also added two paragraphs:

"*Air quality modelling is a key aspect of APHH-Beijing. This involves multiple models from regional (e.g., WRF-Chem, UKCA, NAQPMS) to urban (e.g., CMAQ) and to street scales (ADMS). This section aims to provide an example comparison of model simulated pollutant concentrations against APHH-Beijing measurements made at IAP (Figure 16) to demonstrate model capabilities. Specific modelling work will be published in the special issue.*

Figure 16 shows that the magnitude and variation of wintertime PM2.5 concentrations are reproduced very well by NAQPMS during November, although there are some weakness in capturing the highest PM2.5 levels during the haze events at the end of November and start of December. This is partly due to the representation of local meteorological features during this period, and PM2.5 concentrations during the major haze episode on 4 December are much more similar to those measured at Pinggu than at IAP (see Figure 2). The diurnal variations in O3 during the summertime are reproduced relatively well by UKCA, which captures the rapid daytime formation of O3 and strong nighttime removal. The very highest levels of daytime O3 are underestimated with the model, particularly during the episode at the end of May. However, there is a strong local contribution to this as evident from the lower concentrations measured at Pinggu, and these local differences are not fully resolved with the model. Despite this, the day-on-day build-up of daytime O3 during the periods of 22-27 May and 11-16 June is captured well, and demonstrates that the model reproduces the synoptic drivers of local O3 formation well."

Introduction to Special Issue - In-depth study of air pollution sources and processes within Beijing and its surrounding region (APHH-Beijing)

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Abstract. APHH-Beijing (Atmospheric Pollution and Human Health in a Chinese Megacity) is an international collaborative project to examine the emissions, processes and health effects of air pollution in Beijing. The four research themes of APHH-China are: (1) sources and emissions of urban atmospheric pollution; (2) processes affecting urban atmospheric pollution; (3) exposure science and impacts on health; and (4) interventions and solutions to reduce health impacts. Themes 1 and 2 are closely integrated and support Theme 3, while Themes 1-3 provide scientific data for Theme 4 on the development of cost-effective air pollution mitigation solutions. ~~A key activity within APHH-Beijing was the two month-long intensive field campaigns at two sites: (i) central Beijing, and (ii) rural Pinggu. The coordinated campaigns provided observations of the atmospheric chemistry and physics in and around Beijing during 10 November – 10 December 2016 and 15 May- 22 June 2017. The campaigns were complemented by numerical air quality modelling and air quality and meteorology data at the 12 national monitoring stations in Beijing.~~ This introduction paper provides an ~~overview~~ introduction of (i) the rationale of APHH-Beijing programme, (ii) the measurement and modelling activities performed as part of it in Beijing, and (iii) the air quality and meteorological conditions during the two field campaigns. A key activity within APHH-Beijing was the two month-long intensive field campaigns at two sites: (i) central Beijing, and (ii) rural Pinggu. The coordinated campaigns provided observations of the atmospheric chemistry and physics in and around Beijing during 10 November – 10 December 2016 (winter) and ~~15~~21 May- 22 June 2017 (summer). The campaigns were complemented by numerical air quality modelling and air quality and meteorology data at the 12 national monitoring stations in Beijing. The winter campaign was characterised by several high $PM_{2.5}$ pollution events with peak hourly concentrations at the urban site ranging up to 498 μg m^{-3}, whereas the summer experienced events of high ozone concentrations with the highest hourly average up to 176 ppb. Air quality was generally poor during the winter campaign with an average $PM_{2.5}$

concentration of 96 µg m^{-3}, but less severe than in the same period in 2015. Synoptic scale meteorological analysis suggests that the greater stagnation and weak southerly circulation in November/December 2016 ~~may have~~ contributed to the poor air quality during all haze events detected. $PM_{2.5}$ levels were relatively low during the summer campaign with the highest daily concentration of only 79 µg m^{-3}, matching the cleanest periods over the previous five years. ~~The winter campaign was characterized by high $PM_{2.5}$ pollution events whereas the summer experienced high ozone pollution events. Air quality was poor during the winter campaign, but less severe than in the same period in 2015 when there were a number of major pollution episodes. $PM_{2.5}$ levels were relatively low during the summer period, matching the cleanest periods over the previous five years. Synoptic scale meteorological analysis suggests that the greater stagnation and weak southerly circulation in November/December 2016 may have contributed to the poor air quality.~~

1. INTRODUCTION

Air pollution is one of the largest environmental risks. It is estimated that air pollution has led to 7 million premature deaths per year globally (WHO, 2016a, b) and over a million in China (GBD MAPS Working Group, 2016). Air pollution also has significant impact on the healthcare system and ecosystems, which cost about 0.3% of global GDP (OECD, 2016). Air pollution related sickness also reduced productivity and severe hazes lead to closure of transport systems, causing additional damage to the economy. Total economic losses related to China's $PM_{2.5}$ (particulate matter with aerodynamic diameter equal to or less than 2.5 µm) pollution in 2007 amounted to 346 billion Yuan (£39 billions, approximately 1.1% of the national GDP) based on the number of affected Chinese employees whose work time in years was reduced because of mortality, hospital admissions and outpatient visits (Xia et al., 2016).

Although air pollution in developed megacities sometimes breaks country specific limits and WHO guidelines, traditional London or Los Angeles type smogs which occurred in the early and mid-20th centuries are rare in developing cities to the same extent. In the developing countries however, the rush to industrialisation and rapid growth in vehicle populations have led to serious air pollution problems that are more complex than the London or Los Angeles smogs. Air pollution is particularly severe in developing megacities, such as Beijing, where ~~rapid urbanisation has led to a fast increase in pollution emissions~~ pollutants from traditional sources, such as solid fuel combustion are mixed with those from modern vehicles (Guan et al., 2014), on top of regional pollution from industrial and other anthropogenic activities.

Considerable research effort has led to huge progress in understanding the sources and pollution processes in megacities in western countries, e.g., major interdisciplinary and multi-institutional programmes in Mexico City, Paris and London in the last few years (Molina et al., 2010; Beekmann et al., 2015; Bohnenstengel et al., 2014). Air pollution in megacities in developing countries is different to that in other well studied developed megacities, such as Paris, Mexico City and London, in a number of ways including the lack of diesel emissions in the inner city, the use of coal in surrounding rural areas for heating and domestic cooking (Tao et al., 2018), the high emissions of air pollutants in neighbouring provinces (Hebei and Tianjin) and the high oxidising power due to the complex chemistry (Zhang et al., 2009; Li et al., 2017; Lu et al., 2018). This makes Beijing a particularly interesting place to study as it provides an atmospheric environment very different to developed megacities such as London and Paris to investigate urban pollution processes.

Many research programmes were initiated in Beijing to study the air pollution processes since late 1990s. Earlier research programmes (e.g., early 2000) focused on primary emissions of SO_2, NO_2, CO, PM_{10}, volatile organic compounds and then secondary pollutants such as ground-level ozone and secondary fine particles. These researches contributed to the development of air pollution mitigation strategies by the Beijing Municipal government.

Beijing Olympic Games (2008) offered additional incentives to improve air quality and this led to the funding of CAREBEIJING (Campaigns of Air Pollution Research in Megacity Beijing and Surrounding Region) and other major programmes. CAREBEIJING was initiated and organized by Professor Tong Zhu of Peking University, with participation of hundreds of scientist and students from China, USA, Germany, Italy, Japan, and South Korea. The field campaigns were conducted in the summer of 2006, 2007, and 2008, with the objectives to learn the environmental conditions of the region, to identify the processes (transport and transformation) that lead to the impact of the surrounding area on air quality in Beijing, to quantify the impact of the surrounding area on air quality in Beijing, and to formulate policy suggestion for the air quality attainment during the 2008 Beijing Olympic Games. Other major research programmes, initiated since early 2000, aimed to provide scientific basis to deliver air pollution mitigation measures for ensuring a good air quality during the Olympics Games. Measures developed as a result of these programmes successfully reduced the air pollution during the Olympics Games, and provided valuable examples for air pollution control policy-making in other cities (Wang et al., 2010). CARE-BEIJING latter on was extended to CAREBEIJING-NCP (Campaigns of Air Pollution Research in Megacity Beijing and North China Plain), where field campaigns were carried out in the summer of 2013

and 2014 to investigate the transport and transformation processes of air pollutants in megacity Beijing and North China Plain. The results of CAREBEIJING and CAREBEIJIN-NCP have been published in three special issues of Atmospheric Chemistry and Physics (https://www.atmos-chem-phys.net/special_issue198.html) and Journal of Geophysical Research-Atmospheres (https://agupubs.onlinelibrary.wiley.com/doi/toc/10.1002/(ISSN)2169-8996.CARBS1). These large research programmes and numerous discovery science projects significantly enhanced our understanding on the emission, sources and processes of air pollutants in Beijing (Chan and Yao, 2008; Zhu et al., 2012). However, our understanding of sources and emissions of key air pollutants such as $PM_{2.5}$ and ozone and the role of the interactions between physical and chemical processes in the formation of pollution events in the Beijing megacities is still far from being accurate or complete. In addition, none of the abovementioned large programmes are directly linked heath effect studies. ~~Air pollution in megacities in developing countries, in particular in China have been extensively studied, e.g., in CAREBEIJING (e.g., Liu et al., 2012). However, our understanding of sources and emissions of key air pollutants such as $PM_{2.5}$ and ozone plus the interaction of physical and chemical processes in the formation of pollution events in developing megacities is still far from being accurate or complete.~~

~~Beijing's air pollution is different to that in other heavily studied~~ developed ~~megacities, such as Paris, Mexico City and London, in a number of ways including the lack of diesel emissions in the inner city, the use of coal in surrounding rural areas for heating and domestic cooking (Tao et al., 2018), the high emissions of air pollutants in neighbouring provinces (Hebei and Tianjin) and the high oxidising power due to the complex chemistry (Zhang et al., 2009; Li et al., 2017; Lu et al., 2018). This makes Beijing a particularly interesting place to study as it provides an new~~ contrasting atmospheric environment ~~very different to developed megacities such as London and Paris to test our understanding of~~investigate ~~urban pollution processes.~~

Adverse health effect of air pollution is one of the key motivations to control air pollution. Research has shown that air pollution is one of the leading causes of disease burden in China (GBD MAPS Working Group, 2016). Especially, particulate pollution, the leading cause of severe air pollution events in China, has a significant impact on human health and is associated with high mortality (Zhang et al., 2017a), with considerable proportion of this related to cardiorespiratory diseases (namely stroke, ischemic heart disease, and chronic obstructive pulmonary disease) (Yang et al., 2013; Lozano et al., 2013). Despite this increasing evidence base, the adverse health impact of air pollution remains a complex issue. For instance, the risk assessment of disease burden due to air pollution in China relied largely on the studies undertaken

in Europe and North America, which likely over-simplifies estimates due to the difference of race, life style, air pollution settings (Lim et al., 2012). The marked change in air pollution sources and composition between heating and non-heating seasons, and the differences between urban and rural areas may all lead to different biological responses in local residents. However, to date, such comparative investigations are largely lacking. A further limitation of such work is the lack of accurate personal exposure estimates which are crucial in high quality health studies. This may be especially true when considering household air pollution (both indoors and outdoors) from traditional biomass and coal stoves which may not be easily captured by ambient located monitoring instruments (Linn et al., 2001; Brook et al., 2002). To address current uncertainties and challenges it is essential to improve understanding of the health impact of air pollution worldwide, and to develop mitigation measures with limited resources on health services.

To address these issues, the UK Natural Environment Research Council (NERC), in partnership with the National Science Foundation of China (NSFC), UK Medical Research Council (MRC) and UK-China Innovation Newton Fund funded a major joint research programme – Atmospheric Pollution and Human Health in a Chinese Megacity (APHH-Beijing). The APHH-Beijing is an integrated research programme, incorporating capability and strength of UK and China science community~~The APHH programme~~ and is taking a multi-disciplinary approach to investigating (1) sources and emissions of urban atmospheric ~~pollution~~pollutants; (2) processes affecting urban atmospheric pollution; and (3) the exposure and impacts of air pollution on human health. The scientific understanding from these three themes underpin the development of interventions and solutions to improve air quality and reduce health impacts.

This special issue "In-depth study of air pollution sources and processes within Beijing and its surrounding region (APHH-Beijing)" documents the research outcomes of this APHH-Beijing programme, in particular the atmospheric measurement and modelling aspects.

This introduction paper describes the motivation and background~~aims and objectives~~ of APHH-Beijing programme, and presents some of the background air quality and meteorology observations ~~that form the basis of data interpretation for the whole programme~~, particularly during ~~.~~ the two intensive field campaigns. These campaigns form one of the core research activities within APHH-Beijing integrating the different themes / projects. ~~essential for data interpretation in publications.~~ We did not intend to present the key scientific results of APHH-Beijing here as much of the research activities are still ongoing and unpublished. Such information is more suitable to go to an overview paper instead.

2. APHH-BEIJING PROGRAMME OBJECTIVES

The overall aim of APHH-Beijing is to better understand the sources, atmospheric transformations and health impacts of air pollutants in the Beijing megacity and to improve the capability of forecasting air quality and developing cost-effective mitigation measures. Specific objectives include:

- to determine the emission fluxes of key air pollutants and to measure the contributions of different sources, economic sectors and regional transport to air pollution in Beijing
- to improve understanding of the processes by which pollutants are transformed or removed through transport, chemical reactions and photolysis and the rates of formation and conversion of particulate matter via atmospheric reactions
- to improve understanding on how the detailed properties of particulate matter evolve and can influence their physical properties and behaviour in the atmosphere and elucidate the mechanisms whereby those properties may interact and feedback on urban scale and regional meteorology
- to exploit new satellite observations and regional models to place the *in-situ* campaigns into a wider context
- to determine the exposure of Beijing inhabitants to key health related pollutants using personal air pollution monitors and assess the association between air pollution exposure and key cardiopulmonary measures
- to determine the contribution of specific activities, environments and pollution sources to the personal exposure of the Beijing population to air pollutants derived from outdoor sources
- to enhance our understanding of the health effects in susceptible individuals over time periods when there are large fluctuations in pollutants compared with normal controls, and to identify health outcomes of air pollution.
- to estimate economic loss due to both physical and mental impacts of air pollution and examine how Beijing can improve its air quality more cost effectively

3. RESEARCH THEMES AND INTEGRATION WITHIN THE APHH-BEIJING PROGRAMME

The APHH-Beijing programme has four themes to address specific objectives (Section 2).

3.1 Research Themes

3.1.1 Sources and emissions

This topic is addressed by the AIRPOLL-Beijing (Source and Emissions of Air Pollutants in Beijing) project. AIRPOLL aimed to quantify the emission fluxes of key air pollutants in Beijing and the

contributions of different sources, economic sectors and regional transport to air pollution in Beijing. Several science topics addressed individual issues, which are integrated to achieve the overall aims. The project carried out two major field measurement campaigns jointly with the AIRPRO (The integrated Study of **AIR** Pollution **PRO**cesses in Beijing) and AIRLESS (Effects of **AIR** pollution on cardiopu**L**monary dis**E**a**S**e in urban and peri-urban re**S**idents in Beijing) projects (section 3.1.2 and 3.1.3) using sites within Beijing (at the Institute of Atmospheric Physics (IAP)) and in the local region (the rural Pinggu site – see 4.1 for site information). During winter and summer sampling campaigns, AIRPOLL measured the concentrations of key tracers and reactive species indicative of sources and chemical pathways at the ground sites. AIRPOLL also analysed the vertical concentration profiles measured in conjunction with data from monitoring sites across Beijing.

As Beijing is subject to long-range transport of pollutants from neighbouring regions, a key aim was to differentiate advected pollutants from local emissions. Local sources include road traffic, cooking, burning of fossil fuels by industry and for domestic heating. Secondary pollutants are expected to be largely advected, but the geographic scale of Beijing is sufficient for some formation of secondary pollutants within the city.

During the intensive campaigns, the project measured the fluxes of particulate and gaseous air pollutants from ground-level sources by sampling on a tower at the IAP site, which are being compared with estimates taken from the inventory for Beijing. This was complemented by top-down fluxes inferred from satellite data for nitrogen dioxide, sulphur dioxide and formaldehyde, the latter indicative of VOC oxidation processes (Palmer et al., 2003; Fu et al., 2007). Through these means, the emissions inventory are being tested, allowing revisions which are being incorporated into the atmospheric modelling work.

AIRPOLL also made very detailed on-line and off-line measurements of airborne particles. This included continuous measurements of size distributions from 1 nm to >10 μm diameter. Large molecules and molecular clusters were also measured by high resolution mass spectrometry, with a view to better understanding atmospheric nucleation processes. The project monitored the chemical composition of particles in real time by Aerosol Mass Spectrometry and analysed the time-integrated particle samples off-line for major and minor constituents, including organic molecular markers. AIRPOLL determined the carbon-14 in water soluble organic carbon, water insoluble organic carbon and elemental carbon in selected time-integrated particle samples with an aim to differentiate fossil and non-fossil particulate carbon. These data are being brought together for use in receptor modelling of particulate matter sources, which are compared with other estimates of source contributions to particulate matter concentrations.

Measured ground-level concentrations both from our campaign sites and the Beijing monitoring network, together with source apportionment results~~, and source apportionment~~ are compared with the predictions of a chemistry-transport model and used to provide a clear distinction between advected regional pollution and the impact of local sources. Divergences between measured and modelled pollutant concentrations will be used to provide critical evaluation of emissions inventories, which will be enhanced iteratively with a view to improving knowledge of the sources and emissions of pollutants affecting air quality in Beijing. Data from AIRPOLL-Beijing measurement and modelling work will also contribute to the aims of the AIRPRO project to elucidate the atmospheric physical and chemical processes determining the measured composition.

3.1.2 Atmospheric processes

AIRPRO aims are to study the basic chemical and physical processes controlling gas and aerosol pollution, localised meteorological dynamics, and the links between them within Beijing's atmosphere. Once released to air, atmospheric processing controls how pollutants are subsequently deposited, transformed into secondary pollutants such as O_3 and particulate matter (PM) or transported away from or within the wider Beijing urban area. Previous studies of pollution in Beijing have shown that the interactions of physical conditions, such as the development of temperature inversion in the atmosphere, and chemical processes, e.g., formation of secondary pollutants, such as aerosol particles and ozone that modulate the most severe air quality events.~~Previous studies of pollution in Beijing have shown that it is often perturbation of the physicochemical and dynamic atmospheric conditions that modulate the most severe air quality events, rather than changes in emissions, for example during the development of stable inversions or periods of strong photochemistry.~~ Central to the project were the intensive *in situ* measurements at the IAP meteorological tower (325 m) in Beijing during November-December 2016 and May-June 2017. We made comprehensive and detailed local observations of both primary emitted chemicals and particles, radical intermediates and secondary products, for periods of contrasting local and regional emissions, solar insolation and air temperature. These data allow the performance of local and regional models of air pollution to be robustly tested, both for final regulated pollutant outcomes and at a more mechanistic level.

The observations collected with instruments from multiple Chinese and UK research groups included complementary measurements of key precursor trace gases such as NO_x, HONO, SO_2, CO, O_3, VOCs and SVOCs, gas phase radicals such as OH, HO_2, RO_2, and NO_3, and PM including chemical (both on-line and offline analyses), biological, physical and optical properties. Through multiple co-located surface

measurements, there was both instrumental redundancy (e.g. for equipment failures) and capacity to evaluate through inter-comparison some hard-to-measure atmospheric gases such as OH, HO_2, N_2O_5, HCHO and other oxygenated VOCs. The project determined the local *in situ* chemical processing of air pollution in the contrasting winter/summertime periods alongside overall atmospheric reactivity, both day and at night, through a combination of modelling and proxy measurements such as measured ozone production efficiency and OH reactivity.

The IAP tower is critical~~-~~ as it allowed vertical profiles of key pollutants up to 320 m to be obtained and, with additional remote sensing of composition and meteorology, provided insight into boundary layer stability and evolution over the diurnal cycle. Quantification of shallow mixed layers proved to be vital for explaining local surface *in situ* chemical processing and also street level concentrations of relevance to exposure. The potentially significant vertical gradients anticipated in some chemicals and PM properties were further quantified using instruments installed on the tall tower and via profiling gondola measurements. The combined datasets, surface and profiles, provide the basis for evaluation of model performance, and notably comparisons for those intermediates that provide indicators of whether secondary pollution production is being correctly simulated.

3.1.3 Health effects

This theme is addressed by AIRLESS and APIC-ESTEE (Air Pollution Impacts on Cardiopulmonary Disease in Beijing: An integrated study of Exposure Science, Toxicogenomics and Environmental Epidemiology) projects.

AIRLESS aimed to advance air quality and health research in ~~China~~ Beijing by bringing together two fields of research that have made rapid advancements in recent years: measurements of a wide range of pulmonary and cardiovascular biomarkers in a panel study and personal monitoring of multiple air pollutants with high spatio-temporal resolution by sensor technology. AIRLESS is also benefiting from the use of an extensive range of pollution metrics and source apportionment results ~~collected~~ from ~~in~~ the Themes 1 and 2 projects. These data are being compared with our personal air quality assessments and be used to further understanding of the nature of the air pollution exposures of residents and how this relates to their health status. The APIC-ESTEE study is examining different aspects of air pollution exposure and health, including population studies and toxicology. One aspect of APIC-ESTEE is investigating the relationship between ambient air pollution and personal exposures, and the impacts of both ambient and personal exposures on subclinical health outcomes. Another part of the study is investigating the real-world exposure-reduction and health impact potential of face-masks, a commonly

used personal level intervention seen in Beijing. APIC-ESTEE also carried out laboratory toxicology studies to investigate the toxic mechanisms of PM, and a cohort of mothers and children were recruited to investigate relationships between pre-natal air pollution exposures and birth and infancy outcomes.

3.1.4 Solutions

This theme is addressed by INHANCE (Integrated assessment of the emission-health-socioeconomics nexus and air pollution mitigation solutions and interventions in Beijing) project. In recognition of the health and socio-economic issues associated with air pollution, China's State Council authorized a 1.75 trillion Yuan investment package: the Air Pollution Prevention Plan in 2013. INHANCE quantitatively evaluated the performance of China's current air pollution policies wherein the effectiveness of current anti-air pollution measures. INHANCE not only considered physical and mental health impact, direct economic impact, but also the cascading indirect economic losses occurred through inter-industrial and inter-regional linkages on the supply side of the economy. INHANCE established and evaluated interactive relationships among exposure, vulnerability, impact on health, implications for industry and economic consequences.

INHANCE compared and qualitatively assessed air quality policies between Beijing and other cities; undertook policy performance assessment modelling; utilised techno-economic inventories for anti-pollution measures to conduct micro cost-benefit analysis of new policies; measured health and macroeconomic costs and benefits in mitigating air pollution, and; transformed evidence generated into practical emission alleviation pathways. On these bases, INHANCE will deliver recommendations regarding integrated policy design and an assessment for policy cost-effectiveness.

3.2 Integration Between the Themes

The APHH-Beijing programme is highly integrated to ensure the biggest possible scientific and policy impacts. One of the most significant integration activities between the different themes is the coordinated joint field campaigns at an urban and a rural site in Beijing for Theme 1, 2 and 3 to fully exploit the complementary measurements and expertise by different research groups, which is described in the following sections. Theme 1 & 2 are closely related and in many senses inseparable. For example, our knowledge of the sources and emissions is essential to interpret the processes while knowledge on the atmospheric physical and chemical processes will help us to more accurately quantify the source emissions, both via actual flux-based measurements and model evaluation of the emission inventories.

To ensure integration Themes 1 and 2 co-located their rural site at Pinggu as that was selected for the Theme 3 panel study.

Modelling airborne concentrations of air pollutants within Themes 1 and 2 are fully integrated, primarily via the UKCA (UK Chemistry and Aerosol), NAQPMS (Nested Air Quality Prediction Model System) and GEOS-Chem models. Both models simulate spatial and temporal variations of key air pollutants and will be evaluated using the new observations of pollutant emission fluxes, updated emission inventories, three-dimensional air quality low cost sensor measurements, comprehensive composition and physics measurements, as well as new process understandings generated from the APHH-Beijing programme. Furthermore, Themes 1 and 2 ADMS (Atmospheric Dispersion Modelling System) modelling results for the campaign periods facilitate estimation of population exposure in Theme 3. Outcomes of Themes 1, 2 and 3 provide Theme 4 with a more accurate estimate of pollution costs and help to develop cost-effective air pollution control measures in Beijing.

The third stream of integration activities involves regular APHH-Beijing programme science and stakeholder engagement meetings to stimulate collaboration and knowledge transfer between different themes and stakeholders. Furthermore, sharing of data was made available via a dedicated depository in Centre for Environmental Data Analysis (www.ceda.ac.uk). All data in the depository will be made publically available by the end of 2022.

4. OVERVIEW OF JOINT FIELD CAMPAIGNS

The two intensive campaigns were from 5th November to 10th December 2016 and 15th May to 22nd June 2017. The campaigns were carried out at both urban and rural sites.

4.1 Site Information

The winter campaign has two main sites. The urban site (39.97N, 116.38 E) is located in the Tower Section of Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences; i.e. at the 325 m meteorological tower. The site, between the fourth and third North ring roads of Beijing (Figure 1), is a residential area. Typical of central Beijing, there are various roads nearby. To the south, north and west there are roads about 150 m away. On site there are 2 to 3 floor buildings to the south, east and west of the tower surrounding by small trees and grasses. There is a canal right to the north of the site. Further to the west is a park covered mainly by conifer pine trees (Yuan Dynasty Wall Heritage).

The rural site in Xibaidian village (40.17N, 117.05 E) in north-eastern Beijing, was collocated with the AIRLESS project cohort. Xibaidian village is about 4 km northwest of Pinggu centre, and about 60 km from IAP. There are many similar small villages nearby. The monitoring station and the clinic used an unoccupied house at the north end of the village away from significant local combustion sources. A two-lane road is about 300 m north to the site. With no centralised heating infrastructure available to the local villages' residents mainly use coal and biomass for heating and cooking in individual homes.

In the summer, an additional site was operated in Gucheng (39.2N 115.7E), Dingxing County, Hebei Province. This site, about 120 km to the southwest of central Beijing, is one of the main highly pollutant transport pathways from Hebei province to Beijing via the southwest passage. The site used a meteorological observatory in a farm field. The nearest town is about 10 km to the northeast. The nearest road is 500 m to the north and the nearest village is about 1 km to the west. Several villages are located around the site.

In addition to the two highly instrumented urban and rural (Pinggu) sites, 21 SNAQ (Sensor Network for Air Quality) boxes, which measure CO, NO, NO_2, CO_2, O_X, size resolved particulates (0.38-17.4 μm), temperature, relative humidity, wind speed and direction (Popoola et al., 2018), were deployed during the summer and winter campaigns across the urban and rural areas of Beijing to map air pollutant variations (red tags, Figure 1). Six additional SNAQ boxes were deployed at six different heights (8, 32, 102, 160, 260, and 320 m) on the IAP tower from 9-23 November 2016 and 25 January-31 December 2017.

Figure 1 also shows the location of the 12 national air quality monitoring stations. Hourly data of criteria air pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, CO and O_3) from January 2013 to December 2017 from the stations were also obtained from official sources by Tsinghua University. The closest air quality station to the urban IAP site is about 3 km away at the Olympic Park.

4.2 Instrumentation

4.2.1 Urban site

Table 1 lists all instruments deployed during the campaigns at the IAP site. The nine instrument containers were at ground level on the campus grass. Their locations are shown in Figure 1c. Online instruments and high volume samplers were deployed at different heights on the meteorological tower. Most instruments ran during both campaigns. Vertical profiles measurements included HONO during pollution events using baskets attached to the tower. Additional online measurements and offline particulate matter samplers were deployed at ground-level, roof of a two storied building to the west (WB) and in a third-floor

laboratory at the south-end of the campus. In addition, high, medium and low volume samplers were placed on the roof of WB for offline characterization and source apportionment.

4.2.2 Rural sites

At Pinggu, online instruments (Table 3) were run within an air-conditioned room on the ground floor with inlets on top of the building. High-, medium- and low-volume PM samplers were deployed on a newly modified flat-roof of the single storey building.

At Gucheng (summer only), a high volume Digitel sampler and a single particle sampler were set up on a deserted basketball court. An Aethalometer AE33 was located on top of a container at the edge of the basketball court. CO and O_3 were also measured in a nearby container.

5 . AIR QUALITY DURING THE FIELD CAMPAIGNS

5.1 Winter

During the winter sampling campaign the daily average concentration of $PM_{2.5}$ at IAP using Partisol gravimetric measurements was 91.2 μg m^{-3} (Table 4) and 94.0 μg m^{-3} from online FDMS (Filter dynamic measurement system) measurements. The maximum hourly $PM_{2.5}$ concentration was 438 μg m^{-3} (Figure 2). $PM_{2.5}$ concentrations significantly exceeded the both the daily air quality limit of China (75 μg m^{-3}) and WHO (25 μg m^{-3}). During the whole winter campaign period, nearly 50% of the hours had $PM_{2.5}$ mass concentration higher than 75 μg m^{-3} (Figure 2). Online PM_{10} concentration observed at the Olympic Park national air quality monitoring station was up to 560 μg m^{-3} during the campaign with an average of 130.6 μg m^{-3}. Average concentrations of NO_2, O_3, SO_2 and CO were 69.7 ± 33.3, 16.4 ± 17.0 and 14.9 ± 11.1 μg m^{-3} and 1.53 ± 1.02 mg m^{-3}, respectively (Table 4). Most of the criteria pollutants showed a similar temporal pattern (Figure 2), except O_3.

The daily average concentration of $PM_{2.5}$ was 99.7 μg m^{-3} at Pinggu (Table 4; based on Partisol gravimetric measurement) but as high as 114.0 μg m^{-3} from the BAM measurement. The maximum hourly $PM_{2.5}$ concentration was 617 μg m^{-3} (Figure 2). Similarly to IAP, nearly 50% of the hours had $PM_{2.5}$ mass concentrations greater than 75 μg m^{-3}. Average concentrations of NO_2, O_3, SO_2 and CO are 46.4 ± 25.5, 22.3 ± 22.2, and 15.4 ± 6.7 μg m^{-3} and 1.47 ± 1.17 mg m^{-3} (Table 4). $PM_{2.5}$ was slightly higher at the rural site but NO, CO and SO_2 were comparable between the two sites. $PM_{2.5}$ and O_3 each had similar temporal patterns at the urban and rural sites (Figure 2), indicating a synoptic scale meteorological impact. The larger difference in the temporal variation of NO, NO_2 and SO_2 may reflect the varying contribution of more local sources. Large differences in temporal patterns of air pollutants were found on 4 December 2016 when $PM_{2.5}$, SO_2 and NO concentrations were much higher at Pinggu than at IAP.

Diurnal cycles of particles, NO_2 and CO showed no distinct peak but an increment during the nighttime, suggesting the possible impact of boundary layer and/or anthropogenic emissions in winter (Figure 3). The peak NO levels at 7 am are likely caused by the morning rush hour road traffic. $PM_{2.5}$ concentration increased sharply from 6 pm at Pinggu (not shown), suggesting important local emissions, likely domestic heating and cooking. SO_2 and O_3 had their highest levels in mid-morning or at noon (Figure 3).

Variations of particles, NO_x and SO_2 show that higher levels of these pollutants when air masses were from the south or southwest (Figure 4), indicating it was impacted by regional transport. All pollutants, except O_3, had higher mass concentrations when wind speeds were low, suggesting a local source. The NO wind rose suggests a strong local source with little contribution from long-range transport. The O_3 concentration was higher during northerlies and when the concentrations of other pollutants such as NO_x and $PM_{2.5}$ were lower (Figure 4).

SNAQ box measurements at six levels (8 to 320 m) during the winter campaign (Figure 5) have similar overall temporal patterns of CO and NO to that measured by standard gas analyser (Figure 2). In most cases, the air pollutant levels are similar at different levels of the tower. There are notable differences in NO, CO and CO_2 on 11, 12 and 16 / 17 November, which suggests that the mixed layer height was low (e.g., <150 m). Interestingly, the O_x (NO_2 + O_3) levels are relatively homogeneous across the different levels. These measurements have implications on the role atmospheric chemistry play in transformation of species in the boundary layer, and the measurements also provide useful information that confirm mixed layer height determinations from independent methods such as the ceilometer.

According to the meteorological standards (QX/T113-2010), haze is defined as: i) visibility < 10 km at relative humidity (RH) <80%; or ii) if RH is between 80 and 95%, visibility < 10 km and $PM_{2.5}$ > 75 μg m^{-3}. During the winter campaign 640 of the 1633 h were classified as haze using visibility data from Beijing Capital Airport (Figure 6); within the haze hours 75% had $PM_{2.5}$ greater than 75 μg m^{-3} (Area A, Figure 6) and the rest had a visibility less than 10 km but with a RH <80% (Area B, Figure 6).

Characteristics of five major haze events during the winter campaign (Figure 2) include that $PM_{2.5}$, NO_2, SO_2 and CO had similar trends but O_3 levels dropped to very low concentration (<2 ppb). The events are defined in Table 2.

5.2 Summer

Concentrations of air pollutants excluding ozone during the summer campaign were much lower than in winter (Figure 7, Table 4). Average daily concentration of $PM_{2.5}$ and PM_{10} at IAP were 31.4 ± 14.7 and 74.9 ± 29.3 μg m^{-3} (based on gravimetric method), respectively. These levels were slightly higher than at Pinggu (27.8 ± 13.3 and 62.9 ± 29.3 μg m^{-3}). Concentrations of ozone were four to five times higher during the summer campaigns (106.9 ± 71.6 μg m^{-33} at IAP, and 91.8 ± 62.7 μg m^{-33} at Pinggu) than in the winter campaign. Average concentration of NO_2, SO_2 and CO are 41.3 ± 23.5 and 6.3 ± 6.8 μg $m^{-3}$and 0.61 ± 0.32 ,g m^{-3}at IAP (Table 4). The concentration of NO_2 and CO were lower at Pinggu while that of SO_2 was similar. Most of the criteria pollutants showed a similar temporal pattern (Figure 2), except O_3.

Diurnal patterns of NO, NO_2, and CO at IAP showed a distinct peak in the early morning, suggesting the contribution of traffic emissions (Figure 7). O_3 and O_x concentration peaked in mid-afternoon.

The IAP $PM_{2.5}$ wind rose suggests both local and regional sources (from the south and south-east direction) impact the site (Figure 4). Unlike winter, high ozone concentrations occur during southerlies to southwesterlies, suggesting a regional source of this pollutant. NO and NO_x were largely from local sources during the summer campaign.

Characteristics of two minor haze events (IAP) during the summer campaign (Figure 7) are shown in Table 2.

5.3 Air quality in Wider Beijing Megacity During the Field Campaigns

Average concentrations of air pollutants ($PM_{2.5}$, PM_{10}, NO_2, CO, SO_2 and O_3) at IAP and Pinggu during the two field campaigns were similar to long term averages for these times of year at the 12 national air quality monitoring sites for 2013-2017 (Table 4).

To assess if the IAP air quality is broadly representative of the wider Beijing megacity, variables are correlated with the 12 national air quality station data (Figure 8). A high correlation occurs with $PM_{2.5}$

across all sites except the rural background air quality station at Ming Tombs; PM_{10}, CO and NO_2 at the urban sites are highly correlated but not with the rural and suburban sites suggesting a more local source for these pollutants, comparing to $PM_{2.5}$ and O_3; SO_2 between sites have lower correlation comparing to all other pollutants. The particularly high correlation of $PM_{2.5}$ and O_3 across almost all sites indicates a regional pollution phenomenon for the two pollutants. These results suggested that the air quality at the IAP urban site was broadly consistent with those at the other urban sites.

In general, $PM_{2.5}$ mass concentrations are similar at all the urban sites including IAP but higher than at the suburban and rural background national monitoring site (Ming Tombs, G2) (Figure 9). The Pinggu rural site in this study, has high $PM_{2.5}$ pollution in the winter campaign but has the lowest concentrations during the summer campaign. This suggests that local anthropogenic sources have a major impact on $PM_{2.5}$ at this site during the winter campaigns. Source apportionment results, notably high time resolution data are being used to explore this.

The closest national air quality station (Olympic Park, or Aotizhongxin in Chinese Pingyin) to IAP has highly correlated $PM_{2.5}$ concentration. This suggests that national air quality stations are of sufficient quality to provide valuable information on the spatial and temporal variation of key pollutants to supplement campaign measurements.

Table 4 show the IAP concentrations data for all air quality variables are very close to the 12 national air quality monitoring stations mean. This lends further confidence that the chosen urban site represented well the overall pollution in the Beijing megacity.

6. SYNOPTIC SCALE METEOROLOGY DURING THE FIELD CAMPAIGNS

Given the importance of horizontal advection and wet deposition to air quality in Beijing, the synoptic circulation patterns are clearly important (Miao et al., 2017; Wu et al., 2017; Zhang et al., 2012). To provide the synoptic context of the APHH-China observations, the daily mesoscale flow patterns are classified (Section 6.1) and put into context using a 30-year climatology (Section 6.2).

6.1 Synoptic Circulation Types

Circulation types (CT) are classified using the classification software by the COST Action 733 "Harmonisation and Applications of Weather Type Classifications for European regions" (Philipp et al., 2010) with (ECMWF Re-Analysis) ERA-Interim 6-h 925 hPa geopotential reanalysis data (Dee et al.,

2011) at its native 0.75° spatial resolution for the domain of interest (103-129° E, 31 - 49° N) centred on Beijing (40° N, 116.5° E) covering the period 1988-2017. ERA-Interim 10 m U and V wind components are used to facilitate interpretation of the flow patterns. Of the COST733 methods (Huth et al., 2008; Philipp et al., 2010, 2016; Tveito and Huth, 2016) two are used: T-Mode PCA (Principal Component Analysis) and SANDRA (Simulated Annealing And Diversified RAndomization clustering). The former have been used in Beijing previously (e.g. Miao et al., 2017; Zhang et al., 2012). The latter is considered to perform well in clustering pressure fields and discriminating environmental variables (e.g. Demuzere et al., 2011; Philipp et al., 2016). Classification is performed with the number of CTs ranging from 7 to 18. 11 CTs from the SANDRA method are selected (Figure 11; Table 5) to adequately represent the general flow conditions around Beijing during the 30 y climatology period (Beck and Philipp, 2010). The CTs are re-ordered according to the daily median $PM_{2.5}$ concentration observed at the Olympic Park (i.e. Aotizhongxin) (Figures 1 and 12) in 2013-2017 with the predominant CTs estimated from midday-midday, i.e. with a 12 h time lag.

As expected, the CTs that occurred during the two field campaign periods are different (Figures 12 and 13). During the winter field campaign, the most frequent circulation type was CT 10 (25 % of the 6 h periods) ~~and~~ which was often preceded by a period of CT 11 (total 16%). Circulation types 9-11 are associated with air masses that may stagnate over the Beijing urban area (Figure 11). However, CT 9 did not occur ~~in winter (or the summer)~~during either field campaign. CT 1 (accounting~~ed~~ for 16% of the time~~), with~~ and CT 2 (1 %) are associated with the Asian winter monsoon which brings cold and dry air masses to eastern China. North-westerly flow (over Beijing) is driven by high pressure in the west of the domain (Figure 11). After these conditions of stagnation or north-westerly flow, CT 3, 4, 6, and 5 were the most frequent in the winter campaign (12.5, 11.8, 8.3 and 7.6 % of the time, respectively). CTs 3 and 5 are associated with relatively low pressure in the northeast (Sep-May period)~~.~~ while CTs 4 and 6 have a further reduction in atmospheric pressure in the NE. ~~The remaining~~One single 6 h period was classified as CT 7, i.e., ~~which occurs when~~ winds ~~are~~ oriented westward from the Bohai Sea.

During the summer campaign (Figure 12b), the most frequent CTs were 5, 8, 6, 7 (34, 32, 12, 11 % of the time, respectively). CT 8, which did not occur during the winter campaign period, is (like CT 6) associated with the summer monsoon advecting moist, warm air from the South and Southeast (Figure 11). Synoptic flow from the Northwest (~~The other two were~~ CT 1 and 4) is relatively rare (7 and 4 %, respectively)~~.~~ during ~~During~~ spring and summer (Mar-Aug~~,~~) as ~~CT 4~~ winds start to turn over the Yellow Sea, weakening the NW flow over Beijing.

In comparison to the field campaigns, ~~during the period 1988-2017~~ the CT frequencies range from 7.2% (CT 2, 10) to 12.9% (CT 8) during the period 1988-2017 with clear seasonal variations in their occurrence (Figure 13).

6.2 Synoptic circulation and Air Quality

The 11 CTs (Section 6.1) are clearly associated with distinct air quality conditions based on analysis of hourly air quality data for 2013-2017 at one of the national urban air quality stations (G4, Olympic Park, Figures 1 and 12). Relatively lower $PM_{2.5}$ concentrations occur (Figure ~~Figure~~ 13b) under NE flow conditions (CTs 1-5), and higher concentrations during southerly flow (CTs 6-8, 10). The highest $PM_{2.5}$ concentrations occur during the heating season when regional flow shows~~associate with~~ stagnation (CT 9, 11). All haze events during the winter campaign (Figure 12) are dominated by those CTs although CTs with NE flow conditions occurred for short periods within the haze events (e.g. 18/11/2016, 04/12/2-16). Ozone levels are highest during CTs 5-8 (Figure ~~Figure~~ 13c) as these predominate during spring and summer (Figure ~~Figure~~ 13d).

Similarly, the average mixed layer height observed at IAP (Table 1) varies with season and CT type (Figure ~~Figure~~ 13a). In the Oct 2016 – Sept 2017 period (Figure ~~Figure~~ 13e), the relative frequency of CTs differs slightly from the long-term climatology (Figure ~~Figure~~ 13d). In December 2016, clear air advection from the NE (CTs 1-3) was less frequent than in the 30-y climatology. However, stagnation with a weak southerly component (CTs 9 and 11) was more frequent (Figure ~~Figure~~ 13f), thus favouring haze with a large positive (40%) $PM_{2.5}$ anomaly (Figure ~~Figure~~ 14g, cf. 5 y average, 2013-2017). In June 2017, south-north contrasts in geopotential were apparently reduced so CT 6 was 24% less frequent, while CTs 4, 7, and 8 were more frequent. This had minimal effect of $PM_{2.5}$.~~;~~ ~~t~~The slight relative increase in O_3 (by 9.5%, Figure ~~Figure~~ 13g) during June and January might be explained by ~~associated~~ cloud cover differences, which is being investigated.

6.3 Meteorological Conditions During the Field Campaigns

To assess how local-scale flow related to ERA-Interim fields (section 6.1), the link between the coarse gridded data and tower-based sonic anemometer observations is explored based on wind roses (Figure 14). The 30 y climatology (Figure 13a, d) confirms the clear seasonality in wind direction affecting the occurrence of CTs discussed (Sect. 0), i.e. during winter intensive campaign period (5 November – 10 December) north-easterly flow clearly dominates while southerly wind directions are most common during the summer campaign period (15 May – 22 June). The wind roses for winter 2016 and summer

2017 (Figure 14b, e) are slightly nosier, however, indicating similar tendencies as the climatology. The general large-scale patterns are consistent with the in-situ wind measurements (Figure 14c, f). However, a slight diversion towards northerly and south-westerly flow and lower wind speeds occurred in winter and summer (Figures 14c and f), respectively, when compared to the larger scale data (Figures 14b and d). In addition, south-westerly flows were more frequent in winter 2016 (Figures 14b and c) than the 30 year average climatology (Figure 14a), which had the potential to bring more polluted air in the upwind Hebei province to the observation sites in Beijing.

At 102 m, the flow is consistent with northerlies and north-westerlies in the winter campaign and dominantly southerly and easterlies during the summer campaign (Figure 15). The measured hourly mean wind speed, temperature and relative humidity were 3.1 m s^{-1}, 8.3 ^{o}C and 43.8 % in winter, and 3.6 m s, 25 ^{o}C and 46.7 % in summer, respectively. Typical diurnal patterns were observed with higher wind speed and temperature during the day and RH at night. During the winter haze events the 120 m wind speed were low (an average of 1.8 m s^{-1}) and mainly from the south-west direction (Figures 15 and 2).

~~6.~~4~~7.~~ PRELIMINARY AIR QUALITY MODELLING AND POLLUTION CLIMATOLOGY OF THE CAMPAIGN PERIODS

Air quality modelling is a key aspect of APHH-Beijing. This involves multiple models from regional (e.g., WRF-Chem, UKCA, NAQPMS) to urban (e.g., CMAQ, NAQPMS) and to street scales (ADMS). This section aims to provide an example comparison~~s~~ of model simulated pollutant concentrations against APHH-Beijing measurements made at IAP (Figure 16) to demonstrate model capabilities. Specific modelling work will be published in the special issue later.

Figure 16 shows that the magnitude and variation of wintertime $PM_{2.5}$ concentrations are reproduced very well by NAQPMS during November, although there are some weakness in capturing the highest $PM_{2.5}$ levels during the haze events at the end of November and start of December. This is partly due to the representation of local meteorological features during this period, and $PM_{2.5}$ concentrations during the major haze episode on 4 December are much more similar to those measured at Pinggu than at IAP (see Figure 2). The diurnal variations in O_3 during the summertime are reproduced relatively well by UKCA, which captures the rapid daytime formation of O_3 and strong nighttime removal. The very highest levels of daytime O_3 are underestimated with the model, particularly during the episode at the end of May. However, there is a strong local contribution to this as evident from the lower concentrations measured at Pinggu, and these local differences are not fully resolved with the model. Despite this, the day-on-day build-up of daytime O_3 during the periods of 22-27 May and 11-16 June is

captured well, and demonstrates that the model reproduces the synoptic drivers of local O_3 formation well.

On basis of NAQPMS modelling, we also investigated ~~To determine~~ how representative the campaign periods were of the selected seasons in Beijing by comparing~~,~~ ~~pollutant~~ pollutant levels ~~were compared~~ with those from the same period each year over the 2013-2017 period. The NAQPMS model was run for the full 5-year period driven by NCEP meteorology and using temporally varying emissions for a single year that is broadly representative of 2017 conditions. Use of annually invariant emissions permits the effect of differing meteorology on pollutant levels to be assessed. The frequency distribution of $PM_{2.5}$ for each campaign period for each year is shown in Figure ~~16~~17. $PM_{2.5}$ in winter 2016 is very similar in characteristics to that in 2014, and both years show 50% greater PM levels than in 2013 or 2017. However, pollutant levels are substantially lower than in the same period in 2015, when three extended pollution episodes led to period-mean $PM_{2.5}$ that was ~~almost twice~~ significantly ~~as~~ larger. In contrast, the summer period in 2017 was relatively clean, with $PM_{2.5}$ levels very similar to 2015, and about 25% less than in 2013, 2014 or 2016.

8. SUMMARY

The APHH-Beijing is an integrated and multidisciplinary research programme by leading UK and China researchers to (1) quantify sources and emissions of urban atmospheric pollutants; (2) elucidate processes affecting urban atmospheric pollution events; (3) estimate the personal exposure and impacts of air pollution on human health, and (4) develop intervention strategies to improve air quality and reduce health impacts in the Beijing megacity. This introduction paper outlines the motivation of the APHH-Beijing programme as well as provides the background air quality and meteorological conditions that form the basis of data interpretation for the whole programme, particularly during the two intensive field campaigns as a core research activity within the programme.

APHH-Beijing has measured the fluxes of key air pollutants, including NO_x, CO, BC, VOCs and speciated particulate matter, applied a suite of traditional and modern techniques to apportion the sources of particulate matter, determined a wide range of pulmonary and cardiovascular biomarkers linking to direct personal exposure and extensive fixed-station monitoring as well as source apportionment results, and evaluated the effectiveness of Beijing's air pollution control policies using both chemical transport models and novel machine learning techniques. A number of papers have already been published under the APHH-China programme including those in APHH-Beijing special issue (Wang et al., 2019; Pan et al., 2019; Xia et al., 2018; Zhou et al., 2018; Wang et al., 2018; Lyu et al., 2019; Hollaway et al., 2019; Du et al., 2018; Liu et al., 2018a,b; Smith et al., 2018). More papers are being prepared for publication in

this special issue and elsewhere, which will cover (but not limited to) emission fluxes of air pollutants, chemical composition and source apportionment of fine particles, satellite observations of trace gases and aerosols, sources and processes leading to haze events and photochemical smogs, physical and optical properties of aerosol particles, formation processes of secondary aerosols, urban meteorology, feedbacks between haze, photochemistry and meteorology, integrated regional and urban scale modelling, personal exposure to air pollutants and human health effects of air pollution.

DATA DEPOSITORY

http://catalogue.ceda.ac.uk/uuid/7ed9d8a288814b8b85433b0d3fec0300

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AUTHOR CONTRIBUTIONS

ZS drafted the manuscript and is the science coordinator of the APHH-Beijing programme. RMH, KBH, ACL, PQF, TZ, FJK, ML, ZWS, DBG and ST are lead PIs of the five research projects who led the funding applications and the research. They also drafted section 2. TV plotted many of graphs and carried out the data analysis. SK, SG and MD carried out analysis and wrote section 6.1-6.2; and YLW, MH, ZFW and OW carried out modelling and plotted Figure 16 &17. PFQ, JL and ZT led the air quality measurements at the two measurements sites. SY, JL, RED, LR, DL, JA, DB, WJ, LC, LC, HC, TD, FKD,

BZG, JFH, MH, DH, CNH, MH, DSJ, XJJ, RJ, MK, LK, BL, LC, JL, WJL, KDL, GM, MM, GM (Mills), JA, XFW, EN, BO, CP, PIP, OP, CR, CY, FL, JG, JC, FL, LYS, YS, SRT, QQW, WHQ, XMW, ZFW, LW, XFW, ZJW, PHX, FMY, QZ, YLZ and MZ contribute to the field observations, laboratory measurements and / or modelling. ZS, SG, RMH., ZT, JL, OW, JA, JB, WJB, DC, DCC, HC, TD, RD, FKD, PQF, MFG, DBG, JFH, KBH, MH, DH, CNH, MH, XJJ, RJ, MK, FJK, LK, ACL, JL, ML, KL, GM (Mann), GM (McFiggans), MM, PM, EN, FO, PIP, CP, CR, ARR, LYS, GYS, DS (Spracklen), DS (Stevenson), YS, XJW, JFL, BB, QC, ZWS, ST, SRT, XMW, ZFW, LW, ZJW, PHX, QZ, YHZ and MZ contributed to the funding applications, programme meetings and relevant programme research and/or supervision.

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TABLE LEGENDS:

Table 1: Overview of measurements in APHH-Beijing at the urban site.

Table 2: Haze periods during the summer and winter campaign periods.

Table 3: Overview of measurements at the Pinggu site.

Table 4: Average air quality variables at IAP, Pinggu and 12 national monitoring sites (12N) during the field campaigns (10 November – 11 December 2016; and 21 May – 22 Jun 2017). The 12 national sites five-year mean concentrations for same times of the years (12N -5Y) and for the same time of the year (campaign period) (12N-campaign). Data are mean ± s.d. (range).

Table 5: Mean and standard deviation (sd) of climatological conditions in Beijing for each circulation type (CT) for 1988-20 17 from Era Interim data with frequency of the CT during the W (winter) and S (summer) campaigns (% of 6 h periods (p)) compared to A-long- term 1988-2017.

FIGURE LEGENDS

Figure 1: Study area topography (source: googlemap) of Beijing / Tianjing / Hebei region (a) with the rectangle showing enlarged study area; locations of measurement sites (Institute of Atmospheric Physics (IAP)– urban Beijing, Pinggu – rural Beijing; and Gucheng – upwind site in Hebei province), SNAQ box sites (red symbols) and the 12 national air quality monitoring stations (G1 to G12, blue symbols) (b); locations of the 9 containers at IAP (c) – instrumentation at each container is shown in Table 1. The shaded area shows the Beijing buildup area. (Source: a and b - Goggle Map topographic background imagery; c – taken by Siyao Yue from IAP).

G1: Wangshouxigong; G2: Dingling; G3: Dongsi; G4: Tiantan; G5: Nongzhanguan; G6: Guanyuan; G7: Haidianquwanliu; G8: Shunyxicheng; G9: Huairouzhen; G10:Changpingzhen; G11: Aotizhongxin (Olympic Park); G12: Gucheng. Categories: Urban: G1, G3, G4, G5, G6, G7, G8, G11, G12; Suburban: G9, G10; Rural: G2.

Figure 2: Time-series of air quality variables at the urban and rural sites during the winter campaign; Five haze events are indicated (shading).

Figure 3: Diurnal patterns of gaseous pollutants normalized by average concentrations at IAP during winter and summer campaigns. Line shows the mean concentrations and shaded area as 95% confidence interval in the difference in mean concentrations

Figure 4: Air pollutants concentration (colour) with wind direction (angle) and wind speed (m s^{-1}) at IAP during the winter and summer campaigns. Data are hourly in time resolution and were from 10 November to 11 December 2016 (winter) and 21 May to 22 June 2017 (summer). The colour scale is for "weighted.mean" where the mean wind speed/direction bin is multiplied by the bin frequency and divided by total frequency.

Figure 5: Time series of CO_2, CO, NO, O_x (NO_2+O_3) and wind speed at six heights (colour) measured with SNAQ boxes on the IAP tower during the winter intensive field campaign.

Figure 6: Hourly $PM_{2.5}$ mass concentrations versus visibility (at the Beijing Capital Airport) during the winter campaign. Data source: visibility downloaded using R-"worldmet" package: date of last access: 27/02/2018).

Figure 7: Time-series of air quality variables at the urban and rural sites during the summer campaign. Two minor haze events are indicated (shading).

Figure 8: Correlations between the air quality at IAP, PQ and 12 monitoring station around Beijing. Stations G1-G12 (Figure 2) are labelled 01-12, PG = Pinggu.

Figure 9: Spatial distribution of hourly mean concentration of $PM_{2.5}$ in Beijing during two sampling campaigns.

Figure 10: Hourly $PM_{2.5}$ at IAP (roof of a two storied building) and the neighbouring Olympic park national air quality monitoring station during the winter and summer intensive field campaigns.

Figure 11: ERA-Interim (1988-2017) average 925 hPa geopotential with 10 m horizontal wind vector for 11 circulation types classified for Beijing (municipal boundary thin solid line) surroundings (103-129° E, 31 - 49° N) determined with the SANDRA method (COST733 class software). Frequency of occurrence is given in cluster caption. For discussion of conditions associated with each CT see section 6.1.

Figure 12: Time series of circulation types (CTs) during the two field campaigns: (a) winter and (b) summer. The 11 CTs are shown in Figure 11. See text for more description.

Figure 13: Analysis by circulation type (CT; Sect. 0) of: (a) daily maximum mixed layer height (MLH) determined from ALC observations at IAP between November 2016 – June 2017 (analysis method, Kotthaus and Grimmond, 2018b); concentration of (b) $PM_{2.5}$ and (c) O_3 at at the Olympic Park (i.e. Aotizhongxin) in 2013-2017 from the national air quality network; occurrence of CTs in (d) 1988-2017 and (e) Oct 2016 – Sept 2017; (f) anomaly of CT frequency during Oct 2016 – Sept 2017 compared to 30 y climatology; and (g) anomaly of $PM_{2.5}$ and O_3 during Oct 2016 – Sept 2017 compared to 5 y (2013-2017) average (same data as in b, c).

Figure 14: Beijing wind roses: (a, b, d, e) ERA-Interim 10 m horizontal wind (40° N, 116.5° E) and (c, f) sonic anemometer (Table 1) at IAP 320 m agl for (a) 5 November – 10 December in 1988-2017, (d) 15 May – 22 June in 1988-2017, (b, c) 5 November – 10 December 2016, and (e, f) 15 May – 22 June 2017.

Figure 15: Hourly meteorological variables measured at 120 m during the (a) winter and (b) summer campaigns. The shaded areas highlighted the haze periods (Table 3, Figures 2 and 7).

Figure 16: Comparison of observed and modelled pollutant concentrations showing (a) $PM_{2.5}$ concentrations during the winter campaign compared with NAQPMS simulations, and (b) O_3 mixing ratios in summer compared with UKCA simulations.

Figure 17: Frequency distribution of $PM_{2.5}$ in Beijing over the winter (left) and summer (right) campaign periods from the NAQPMS model compared with those from the same periods over the past five years under the same emission conditions.

1398 **Table 1:** Overview of measurements in APHH-Beijing at the urban site.

	Instrument	Measurements	Institute	References
Container 2	FAGE	OH (Chem and Wave)^{X}, HO_2, RO_2	Leeds	Whalley et al. (2010)
	OH reactivity	OH reactivity	Leeds	Stone et al. (2016)
	Spectral radiometer	Photolysis rates	Leeds	Bohn et al. (2016)
	Filter radiometer	$J(O^1D)$	Leeds	Bohn et al. (2016)
	Dew point hygrometer	Water vapour	Leeds	Whalley et al. (2010)
	Davis met station	Wind speed, direction, temp, RH, pressure	Leeds	
	Vaisala CL31 ALC Ceilometer $^{+}$	Cloud-base height, mixing height, attenuated backscatter profiles	Reading	Kotthaus and Grimmond (2018a)
	Personal air monitors (PAMS)	CO, NO, NO_2, PM_1, PM_{10}, $PM_{2.5}$	Cambridge	Moore et al. (2016)
	MicroPEMs	Personal PM exposure	IOM	Sloan et al. 2015
Container 2	DC-GC-FID	C2-C7 VOCs and oVOCs	York	Hopkins et al. (2011)
	GCxGC FID	C6 - C13 VOCs and oVOCs	York	Dunmore et al. (2015)
	TEI 42i	NO	Birmingham	
	Teledyne CAPS	NO_2	York	
	TEI 42c	Total NO_y	York	
	TEI 49i	O_3	York	
	TEI 43i	SO_2	York	
	Sensor box	CO	York	Smith et al. (2017)
	BBCEAS	HONO, NO_3, N_2O_5	Cambridge	Le Breton et al. (2014)
Container 3	LOPAP	HONO	Birmingham	Crilley et al. (2016)
	LIF HCHO	HCHO	Leeds	Cryer et al. 2016
	LOPAP	HONO	IC-CAS	Zhang et al. (2018)
	GC-MS	Organic nitrates	East Anglia	Mills et al. (2016)
	ROS online analyser	Reactive Oxygen Species	Cambridge	Wragg et al. (2016)
Con tain	FAGE	OH (wave)^{x}, HO_2	Peking	Lu et al., 2012

	FAGE	OH (chem)[x]	Peking	Tan et al., 2017
	TEI 42i	NO	Peking	Tan et al., 2017
	Teledyne CAPS	NO2	Peking	
	TEI 42c with Moly converter	NO_2	Peking	
	TEI 49i	O_3	Peking	
	TEI	CO	Peking	
	Spectral radiometer	Photolysis rates	Peking	
	GC-ECD	PAN	Peking	Zhang et al., 2011
	GC-MS	VOCs	Peking	Wang et al., 2015a
Container 5*	H-TDMA/V-TDMA	Hygroscopicity/volatility	Peking	Wu et al., 2013
	SMPS+APS	Particle Number size distribution	Peking	Wu et al., 2016
	Particle size magnifier	Size distribution of < 3nm particles	Peking	Vanhanen et al., 2011
	IGAC-IC	Water-soluble ions	Peking	Yu et al. (2018)
	Xact	Metal	Peking	Yu et al. (2018)
	Sunset OC/EC	EC/OC	Peking	Zhang et al. (2017b)
Container 6	IBBCEAS	HONO, NO_2	AIOFM	Duan et al. (2018)
	CRDS	NO_3 and N_2O_5	AIOFM	Li et al. (2018)
	Nitrate Api-TOF-CIMS	Organics, clusters (HOMs)	Birmingham	Junninen et al. (2010)
	SMPS	Particle size distribution	Birmingham	Shi et al. (1999)
	Particle size magnifier	Size distribution of < 3 nm particles	Birmingham	Vanhanen et al. (2011)
Container 7	Fast NO_x	NO_x fluxes	York	Vaughan et al. (2016)
	AL5002 CO analyser	CO fluxes	York	Gerbig et al. (1999)
	HR-TOF-AMS	Fluxes of PM_1 non-refractory (NR) species	CEH	Nemitz et al. (2008)
	SP2	BC fluxes	Manchester	Liu et al. (2017)
	PTR-TOF-MS	VOC fluxes	GIG Lancaster	Huang et al. (2016)

	SYFT-MS Voice 200 Ultra	VOC fluxes	York	Storer et al. (2014)
Container 8	SMPS3968-APS3321	Particle number size distribution	BNU	Du et al. (2017)
	H/V TDMA	Particle hygroscopicity	BNU	Wang et al. (2017b)
	CCNC-100	CCN	BNU	Wang et al. (2017b)
	PAX (870nm)	Extinction & absorption coefficient	IAP	Xie et al. (2018)
	Ammonia analyzer	NH_3	IAP	Meng et al. (2018)
	Sunset OC/EC analyzer	Online OC/EC	IAP	Zhang et al. (2017b)
Container 9	Iodide FIGAERO-TOF-CIMS	Particle and gas phase molar molecule	Manchester	Le Breton et al. (2018)
	CPMA-SP2	Black carbon mass and mixing state	Manchester	Liu et al. (2017)
	Micro reactor	oVOCs	York	Pang et al. (2014)
Tower ~100 m	QCL NH_3	Ammonia fluxes	CEH	McManus et al. (2010)
	IRGA LiCOR-7500	CO_2 / H_2O flux	CEH	McDermitt et al. (2011)
	DMT UHSAS	Size resolved particle flux (0.06-1 μm)	CEH	Deventer et al. (2015)
	TSI APS3021	Size-resolved particle flux (0.5-25 μm)	CEH	Nemitz et al. (2002)
	TSI CPC3785	Total particle number flux	CEH	Petäjä et al. (2006)
	ROFI	O_3 flux	CEH	Coyle et al. (2009)
	Sonic anemometer R3-50	Turbulence, sensible heat flux	CEH	Högström and Smedman (2004)

	WXT530 weather station	T, P, RH, wind speed & direction, precipitation	CEH	
	2B O_3 analyser	O_3 concentration	CEH	Johnson et al. (2014)
Tower ~120 m	High-vol sampler	$PM_{2.5}$ filter samples	IAP	
	Anderson sampler	Size-resolved PM samples	IAP	
Tower ~260 m	High-vol sampler	$PM_{2.5}$ filter samples	IAP	
	Anderson sampler	Size- resolved PM samples	IAP	
	ACSM	NR PM_1 species	IAP	Sun et al. (2012)
	CAPS-PM-$_{Ext}$ (630nm)	Extinction	IAP	Wang et al. (2015b)
	SMPS 3938	Particle Number size distribution	IAP	Du et al. (2017)
	Gas analyser	CO, O_3 and SO_2	IAP	Zhou et al. (2018)
	Aethalometer AE33	Black carbon	IAP	Xie et al. (2018)
	Single particle sampler	Individual particles	CUMTB	Wang et al. (2018)
Tower and tower basket measurements	SNAQ boxes (x 6 at different heights)	CO, NO, NO_2, SO_2, PM_1, PM_{10}, $PM_{2.5}$	Cambridge	Popoola et al. (2018)
	LOPAP	HONO (3 min avg)	Birmingham	Crilley et al. (2016)
	Spectral radiometer	Photolysis rates	Leeds	Bohn et al. (2016)
	SNAQ	CO, NO, NO_2, SO_2, PM_1, PM_{10}, $PM_{2.5}$	Cambridge	Popoola et al. (2018)
	WIBS	Fluorescent biological aerosol particles (FBAP)	IAP	Yue et al. (2016)
	AE33	BC	IAP	Xie et al. (2018)
	Los Gatos NH_3 Analyzer	NH_3	IAP	Meng et al. (2018)
	PAX	Light scattering / absorption	IAP	Xie et al. (2018)
IAP ground	High-Vol sampler	$PM_{2.5}$ filter samples	Peking	
	4-channel sampler	$PM_{2.5}$ filter samples	Peking	

	High Vol sampler	*High time resolution* $PM_{2.5}$ filter samples	York	
IAP roof/lab	FDMS+Thermo Scientific 1405-DF	Online $PM_{2.5}$ mass conc.	IAP	
	Partisol sampler	$PM_{2.5}$ + $PM_{2.5\text{-}10}$	Birmingham	Taiwo et al. (2014)
	Streaker sampler	Hourly elements in $PM_{2.5}$ and $PM_{2.5\text{-}10}$	Birmingham	Taiwo et al. (2014)
	Digitel High Vol	$PM_{2.5}$ daily	IAP	
	Digitel High Vol	PM_1 - 3 hourly	IAP	
	Andersen sampler	Size resolved PM	IAP	
	WIBS	Fluorescent biological particles	IAP	Yue et al. (2016)
	CAPS-NO_2 Aethalometer	NO_2	IAP	Ge et al. (2013)
	AE33	Black carbon	IAP	Xie et al. (2018)
	CAPS-PM_{SSA} (630nm)	Extinction, Scattering	IAP	Han et al. (2017)
	HR-ToF-AMS	NR-PM species	IAP	Sun et al. (2016)
	SP-AMS	Refractory BC and coated aerosol composition		Wang et al. (2017a)
	Iodide FIGAERO-ToF-CIMS	Particle and gas phase molar molecule	IAP	Zhou et al. (2018)
	Single particle sampler	Individual particles	CUMTB	Wang et al. (2018)

Institution names: AIOFM = Anhui Institute of Fine Optics and Mechanics; BNU = Beijing Normal University; CEH = Centre for Ecology and Hydrology; CUMTB = China University of Mining and Technology (Beijing); GIG = Guangzhou Institute of Geochemistry, Chinese Academy of Sciences; NUIST = Nanjing University of Information Science &Technology; IC-CAS = Institute of Chemistry, Chinese Academy of Sciences

[+] Deployment of instruments both campaigns unless: 10/11/2016 to 25/6/2017

[*] Winter campaign only

[X] OH wave and OH chem refer to the method used to obtain the background signal for the FAGE instruments which are equipped with a scavenger inlet

1410 **Table 2:** Haze periods during the summer and winter campaign periods.

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Event	Time	$PM_{2.5}$ (μg m^{-3})	Visibility (km)
Winter Haze Event 1	11/08 21:00- 11/10 16:00	158 (79 - 229)	4.1 (2.3-8)
Winter Haze Event 2	11/15 21:00- 11/19 08:00	143 (56 - 244)	4.2(0.6-8)
Winter Haze Event 3	11/24 12:00- 11/27 02:00	210 (68-363)	4.2(1.5-8)
Winter Haze Event 4	12/02 16:00- 12/05 02:00	239 (58 -530)	3.9(0.9-8)
Winter Haze Event 5	12/06 09:00- 12/08 10:00	144 (64 -229)	4.6(2.2-8)
Summer Haze Event 1	27/05 12:00 -28/05 13:00	107(62- 163)	6.8(4.5-9)
Summer Haze Event 2	17/06 09:00-18/06 17:00	90.5(60-153.3)	9.3(7-13)

1412 Note: data in parentheses show the range

1413

1414 **Table 3:** Overview of measurements at the Pinggu site.

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Instruments	Measurements	Insitutue	Reference
Thermo gas analysers	$NO_x/SO_2/CO/O_3$	Peking	Liang et al., 2017
BAM 1020	$PM_{2.5}$ mass concentration	Peking	Liang et al., 2017
High vol sampler	$PM_{2.5}$ samples	IAP	Zhao et al., 2018
Medium vol sampler	$PM_{2.5}$ samples	IAP	Zhao et al., 2018
Low vol Andersen sampler	Size resolved PM samples	IAP	Zhao et al., 2018
Partisol sampler	$PM_{2.5}$ samples	Birmingham	Taiwo et al. (2014)
Streaker sampler	Hourly elements in $PM_{2.5}$ and $PM_{2.5-10}$	Birmingham	Taiwo et al. (2014)
High vol sampler	Filters of $PM_{2.5}$; high time resolution	Birmingham	
Four Channel sampler	$PM_{2.5}$ samples	Peking	Liang et al., 2017
Thermo MAAP	Online Black Carbon	Peking	Lin et al., 2011
Sunset OC/EC analyzer	Online OC/EC	Peking	Han et al., 2014
Xact	Hourly metals	Peking	Yu et al. (2018)
TOF-ACSM	NR-chemical composition (summer)	Peking	Sun et al., 2012
Thermo Metone	Meteorological parameters	Peking	Liang et al., 2017
SNAQ	Meteorological parameters	Cambridge	Popoola et al. (2018)
SP-AMS	Individual particle composition	CQIGIT	Chen et al. (2017)
SMPS	Size distribution	Tsinghua	Wang et al., 2009
ACSM	NR-chemical composition (winter)	Tsinghua	Li et al. (2016)

1416 CQIGIT = Chongqing Institute of Green and Intelligence Technology, Chinese Academy of Sciences

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Table 4: Average air quality variables at IAP, Pinggu and 12 national monitoring sites (12N) during the field campaigns (10 November – 11 December 2016; and 21 May – 22 June 2017). The 12 national sites five-year mean concentrations for same times of the years (12N -5Y) and for the same time of the year (campaign period) (12N-campaign). Data are mean ± s.d. (range).

Pollutant[1]	**Winter** *(10 Nov-11 Dec 2016)*				**Summer** *(21 May-22 June 2017)*			
	IAP	PG	12N-5Y	12N - campaign	IAP	PG	12N-5Y	12N-campaign
$PM_{2.5}$ [2]	91.2 ± 63.7 (10.3-239.9)	99.7 ± 77.8 (13.3-294.3)	84.01 ± 89.1 (3.2-593.3)	95.3 ± 79.6 (4.7-408.8)	31.4 ± 14.7 (12.2-78.8)	27.8 ± 13.3 (10.6-70.3)	58.7 ± 40.0 (4.2-250.3)	41.7 ± 22.3 (8.9-134.1)
PM_{10} [2]	130.6 ± 87.0 (20.0-329.2)	121.9 ± 80.4 (10.4-312.1)	112.8 ± 102.2 (5-662.0)	134.5 ± 100.4 (6.0-550.1)	74.9 ± 29.3 (22.5-164.6)	62.9 ± 29.3 (15.1-141.9)	94.6 ± 52.7 (5.0-463.2)	81.9 ± 37.1 (6.0-277.8)
NO_2	69.7 ± 33.3 (10.2-167.3)	46.4 ± 25.5 (2.3-132.4)	57.7 ± 33.9 (3.9-166.4)	66.4 ± 31.3 (7.3-156.6)	41.3 ± 23.5 (9.2-142.9)	29.3 ± 10.3 (9.3-84.0)	40.6 ± 17.9 (8.1-132.4)	37.6 ± 16.2 (12.5-92.8)
SO_2	14.9 ± 11.1 (0.1-50.8)	15.4 ± 6.7 (6.2-44.4)	16.6 ± 16.2 (1.4-112.0)	14.2 ± 9.4 (2.1-51.4)	6.3 ± 6.8 (0.1-38.2)	8.9 ± 4.7 (4.2-41.2)	10.1 ± 10.6 (1.8-82.3)	7.4 ± 6.6 (1.8-64.5)
CO [2]	1.53 ± 1.02 (0.7-5.0)	1.47 ± 1.17 (0.1-6.9)	1.65 ± 1.38 (0.1-9.6)	1.86 ± 1.17 (0.3-5.7)	0.61 ± 0.32 (0.1-2.5)	0.52 ± 0.29 (0.1-2.3)	0.93 ± 0.74 (0.2-8.7)	0.74 ± 0.33 (0.2-2.5)
O_3	16.4 ± 17.0 (0.3-63.3)	22.3 ± 22.2 (2.9-78.0)	21.8 ± 20.5 (1.0-72.9)	17.5 ± 19.2 (2.1-67.4)	106.9 ± 71.6 (2.0-349.3)	91.8 ± 62.7 (0.2-291.4)	100.4 ± 67.8 (2.2-343.5)	110.8 ± 66.5 (3.6-335.9)

[1], Units: μg m^{-3} except CO units: mg m^{-3}

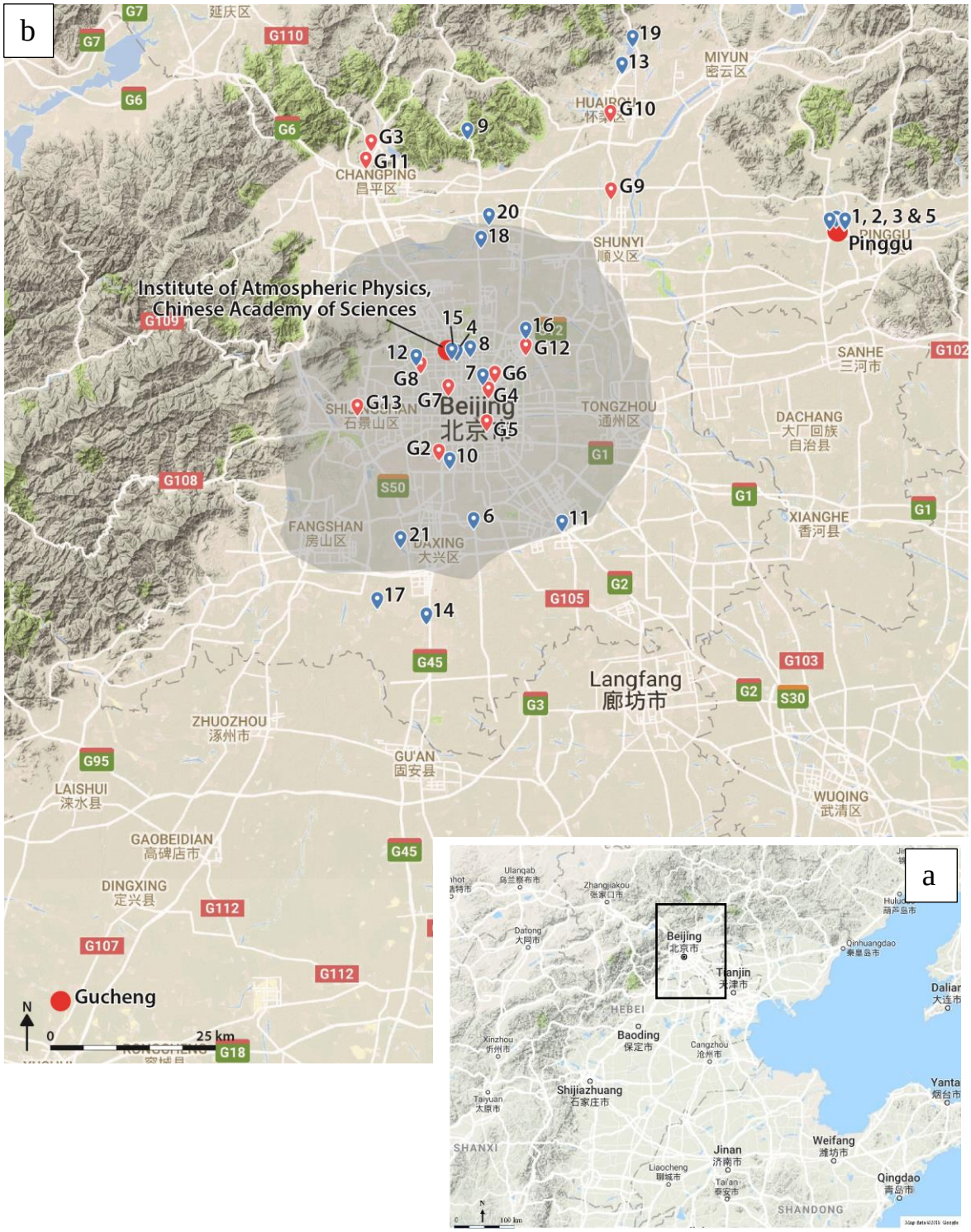
[2], $PM_{2.5}$ and PM_{10} from IAP and Pinggu measured by a gravimetric method; all other data are online measurements hourly mean.

Table 5: Mean and standard deviation (sd) of climatological conditions in Beijing for each circulation type (CT) for 1988-20 17 from Era Interim data with frequency of the CT during the W (winter) and S (summer) campaigns (% of 6 h periods (p)) compared to A- long- term 1988-2017.

CT	Description	WS m s^{-1}	WS_{sd} m s^{-1}	WD °	WD_{sd} °	T2m °C	$T2m_{sd}$ °C	TD2m °C	$TD2m_{sd}$ °C	MSLP hPa	$MSLP_{sd}$ hPa	RH %	RH_{sd} %	Season	Frequency (%) W	S	A
1	H - west of the domain	3.38	1.63	298.3	62.6	0.1	7.1	-12.6	7.9	1026.50	4.14	41	18	Winter monsoon	16	7	9.3
2	H - west of the domain	2.91	1.49	265.9	107.0	-2.8	6.2	-13.8	7.5	1034.34	4.47	45	18	Winter monsoon	1	0	7.2
3	relatively L in NE	3.21	1.65	281.2	71.3	6.8	8.9	-6.4	9.3	1017.77	4.35	43	20	Sep- May	12.5	0	8.3
4	further reduction L (cf. CT3, 5) in NE winds start to turn over Yellow Sea	3.05	1.73	240.1	104.1	19.2	7.5	7.0	10.4	1007.20	3.63	50	24	Mar-Aug Spring - summer	11.8	4	7.8
5	relatively L in NE	2.57	1.37	189.1	125.0	8.2	8.9	-0.9	10.4	1020.82	4.62	57	23	Sep-May	7.6	34	8.3
6	further reduction L (cf. CT3, 5) in NE	2.58	1.32	197.4	87.6	24.6	5.9	14.7	8.0	1000.99	2.96	59	23	Summer monsoon	8.3	12	8.9
7	when winds are oriented westward from the Bohai Sea	2.29	1.12	167.5	100.2	18.9	7.8	10.7	9.5	1012.59	3.61	63	21		1 p	11	10.2
8	like CT 6	2.35	1.11	165.4	75.4	24.0	5.3	15.9	6.8	1006.47	2.69	65	21	Summer monsoon		32	12.9
9	Air mass stagnant over Beijing	2.03	0.94	208.7	107.4	2.1	7.9	-6.2	8.4	1028.66	4.18	58	20			0	9.6
10	Air mass stagnant over Beijing	2.67	1.17	211.1	68.7	14.2	9.4	3.1	10.0	1013.98	3.84	52	22		25	0	7.2

11	Air mass stagnant over Beijing	2.23	0.98	209.1	86.5	8.1	9.4	-0.4	9.6	1021.83	4.06	59	20		16	0	10.3

Note: WS- wind speed, WD wind direction, T2m – 2 m air temperature, TD2m – 2 m dewpoint temperature, MSLP – mean sea level pressure, RH – relative humidity; L – low pressure; H – High pressure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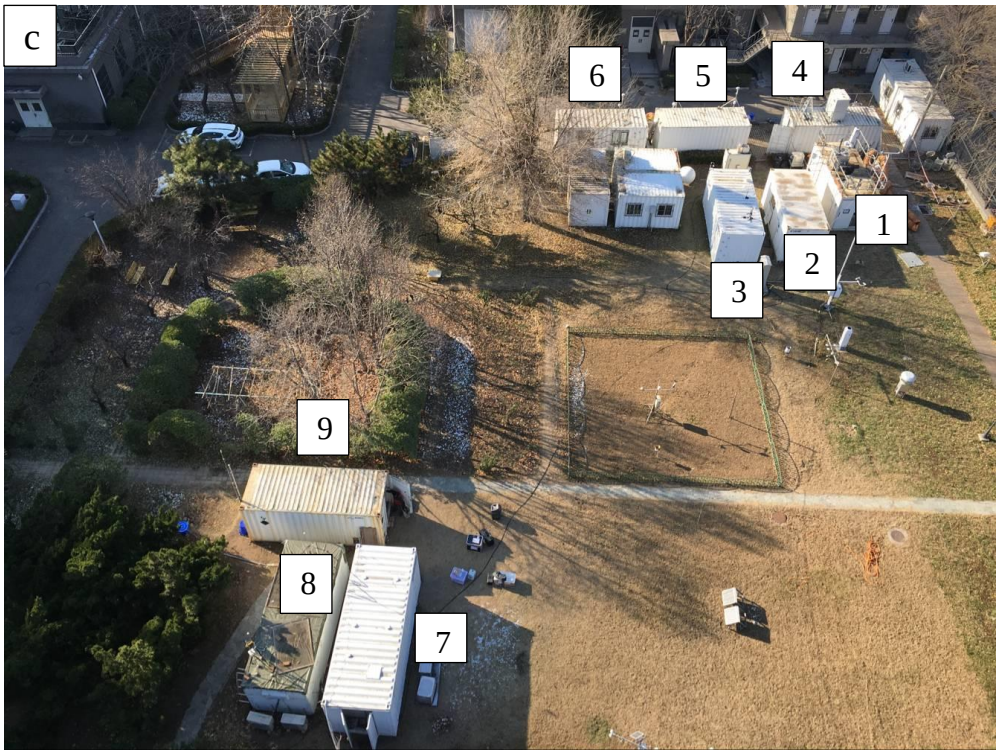


Figure 1: Study area topography (source: googlemap) of Beijing / Tianjing / Hebei region (a) with the rectangle showing enlarged study area; locations of measurement sites (Institute of Atmospheric Physics (IAP)– urban Beijing, Pinggu – rural Beijing; and Gucheng – upwind site in Hebei province), SNAQ box sites (red symbols) and the 12 national air quality monitoring stations (G1 to G12, blue symbols) (b); locations of the 9 containers at IAP (c) – instrumentation at each container is shown in Table 1. The shaded area shows the Beijing buildup area. (Source: a and b - Goggle Map topographic background imagery; c – taken by ~~Siyao Yue~~ Jian Zhao from IAP).

G1: Wangshouxigong; G2: Dingling; G3: Dongsi; G4: Tiantan; G5: Nongzhanguan; G6: Guanyuan; G7: Haidianquwanliu; G8: Shunyxicheng; G9: Huairouzhen; G10:Changpingzhen; G11: Aotizhongxin (Olympic Park); G12: Gucheng. Categories: Urban: G1, G3, G4, G5, G6, G7, G8, G11, G12; Suburban: G9, G10; Rural: G2.

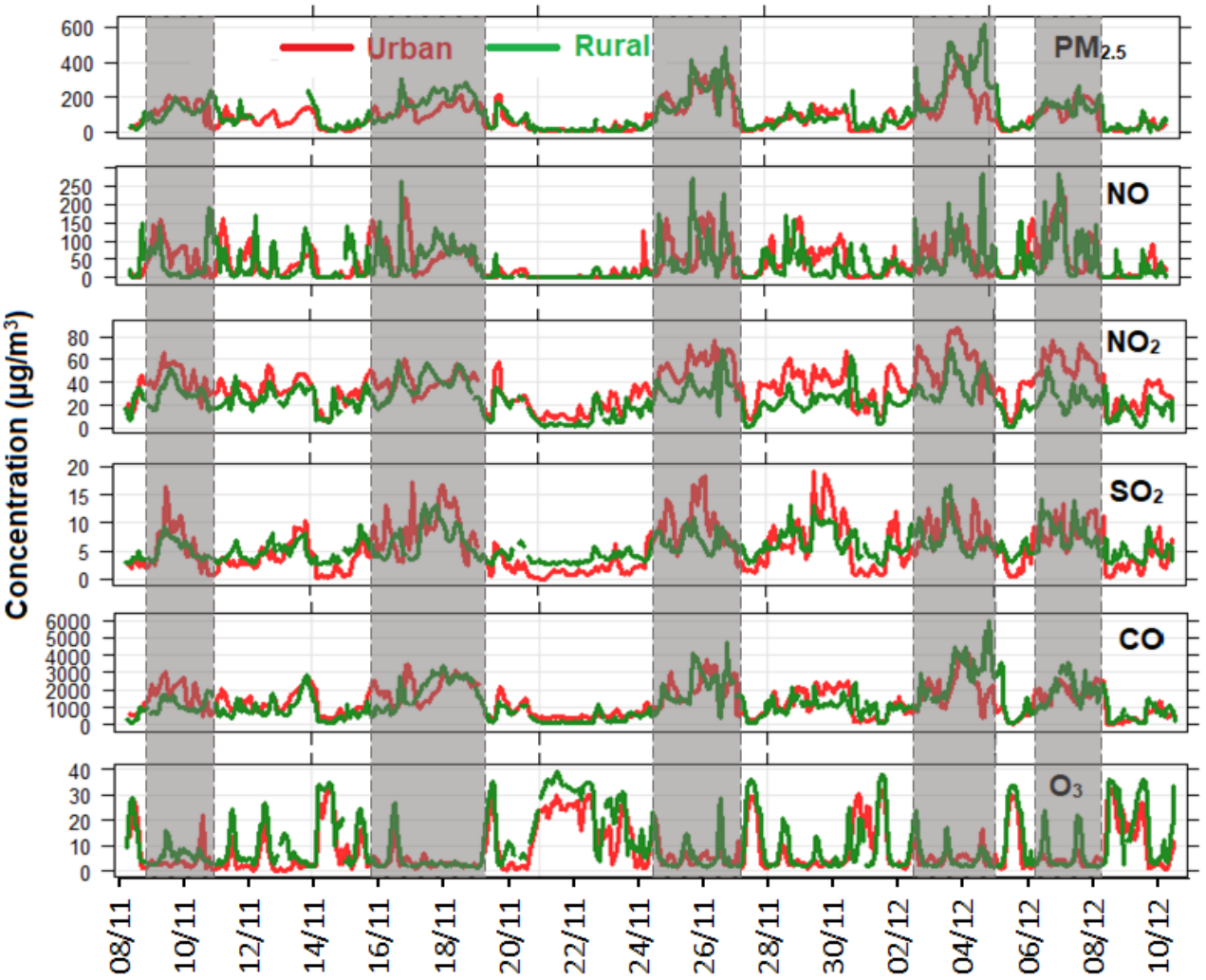


Figure 2: Time-series of air quality variables at the urban and rural sites during the winter campaign; Five haze events are indicated (shading).

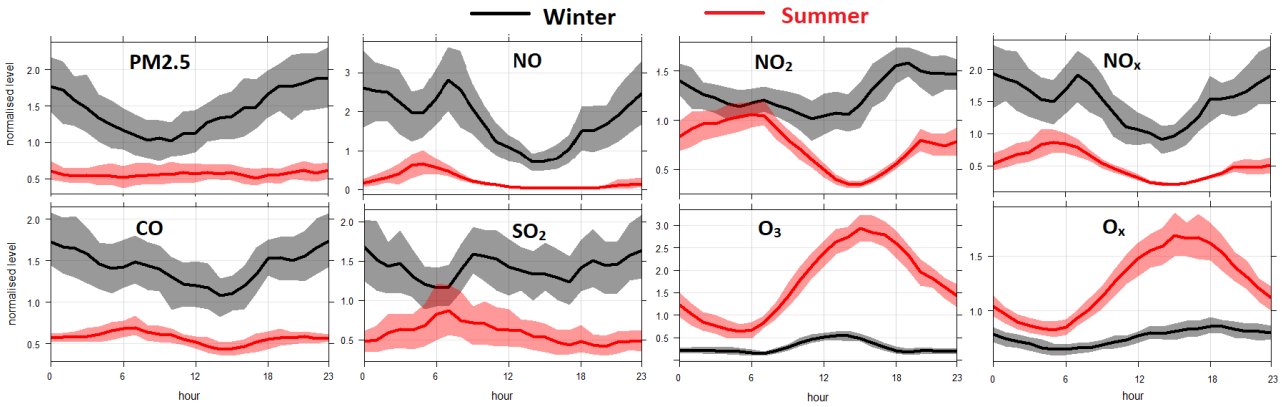


Figure 3: Diurnal patterns of gaseous pollutants normalized by average concentrations at IAP during winter and summer campaigns. Line shows the mean concentrations and shaded area as 95% confidence interval in the difference in mean concentrations.

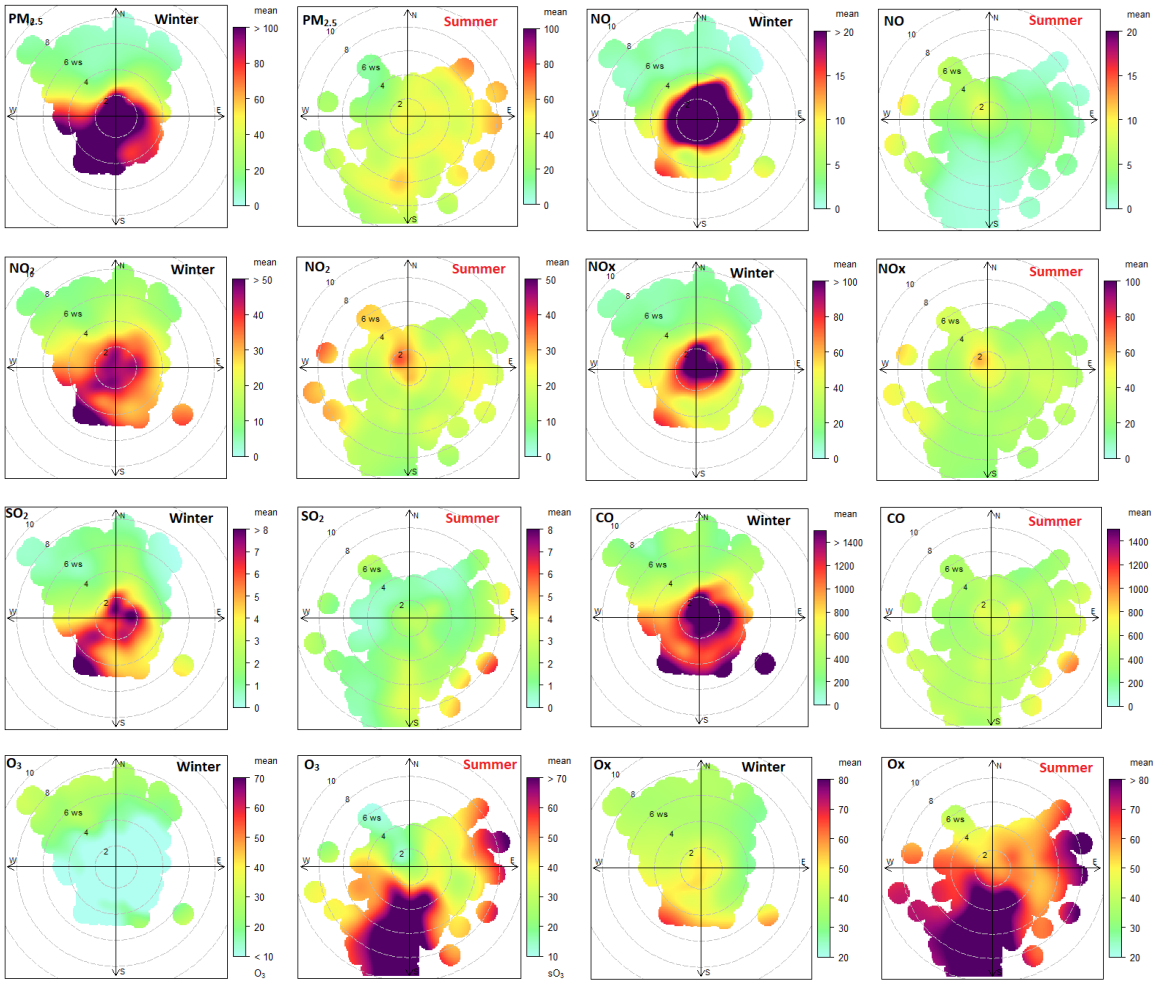
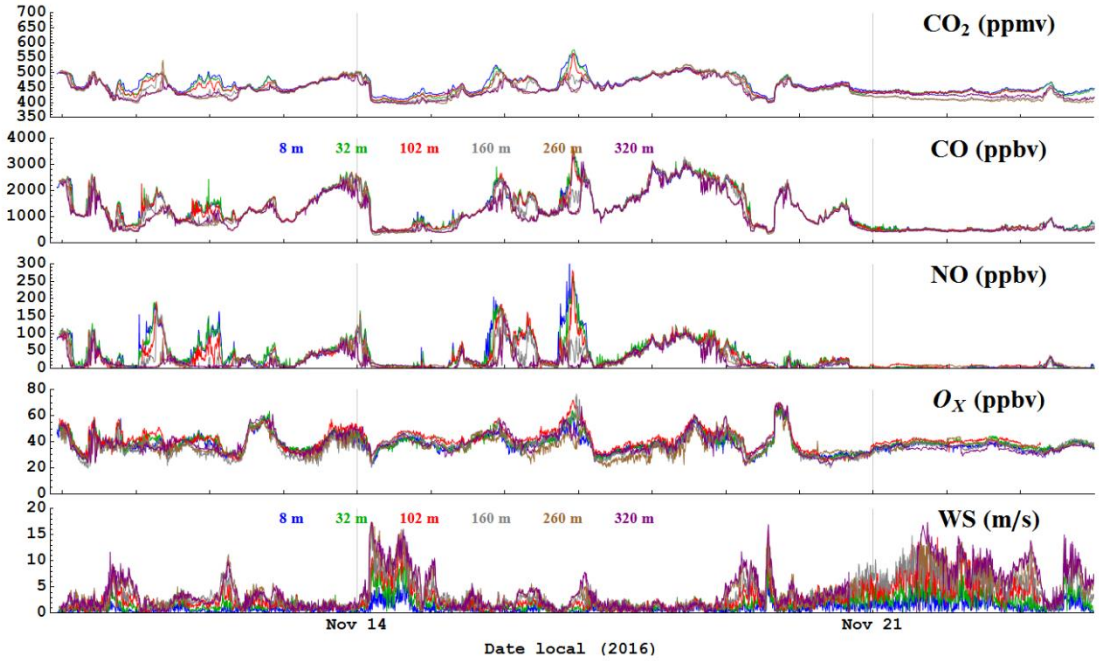


Figure 4: Air pollutants concentration (colour) with wind direction (angle) and wind speed (m s^{-1}) at IAP during the winter and summer campaigns. Data are hourly in time resolution and were from 10 November to 11 December 2016 (winter) and 21 May to 22 June 2017 (summer). The colour scale is for "weighted.mean" where the mean wind speed/direction bin is multiplied by the bin frequency and divided by total frequency.



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1489 **Figure 5:** Time series of CO_2, CO, NO, O_x (NO_2+O_3) and wind speed at six heights (colour)
1490 measured with SNAQ boxes on the IAP tower during the winter intensive field campaign.

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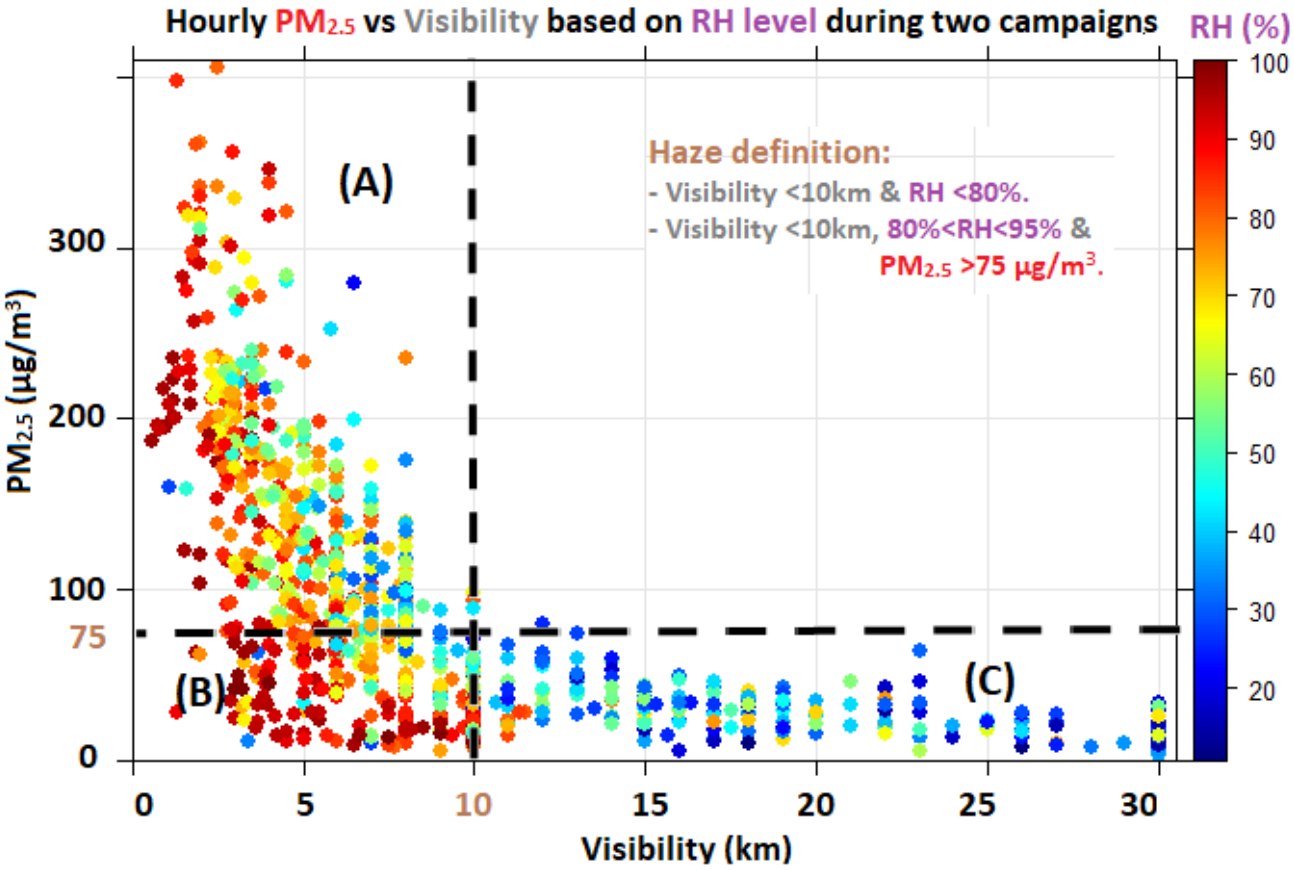

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1494 **Figure 6:** Hourly $PM_{2.5}$ mass concentrations versus visibility (at the Beijing Capital Airport) during
1495 the winter campaign. Data source: visibility downloaded using R-"worldmet" package: date of last
1496 access: 27/02/2018).

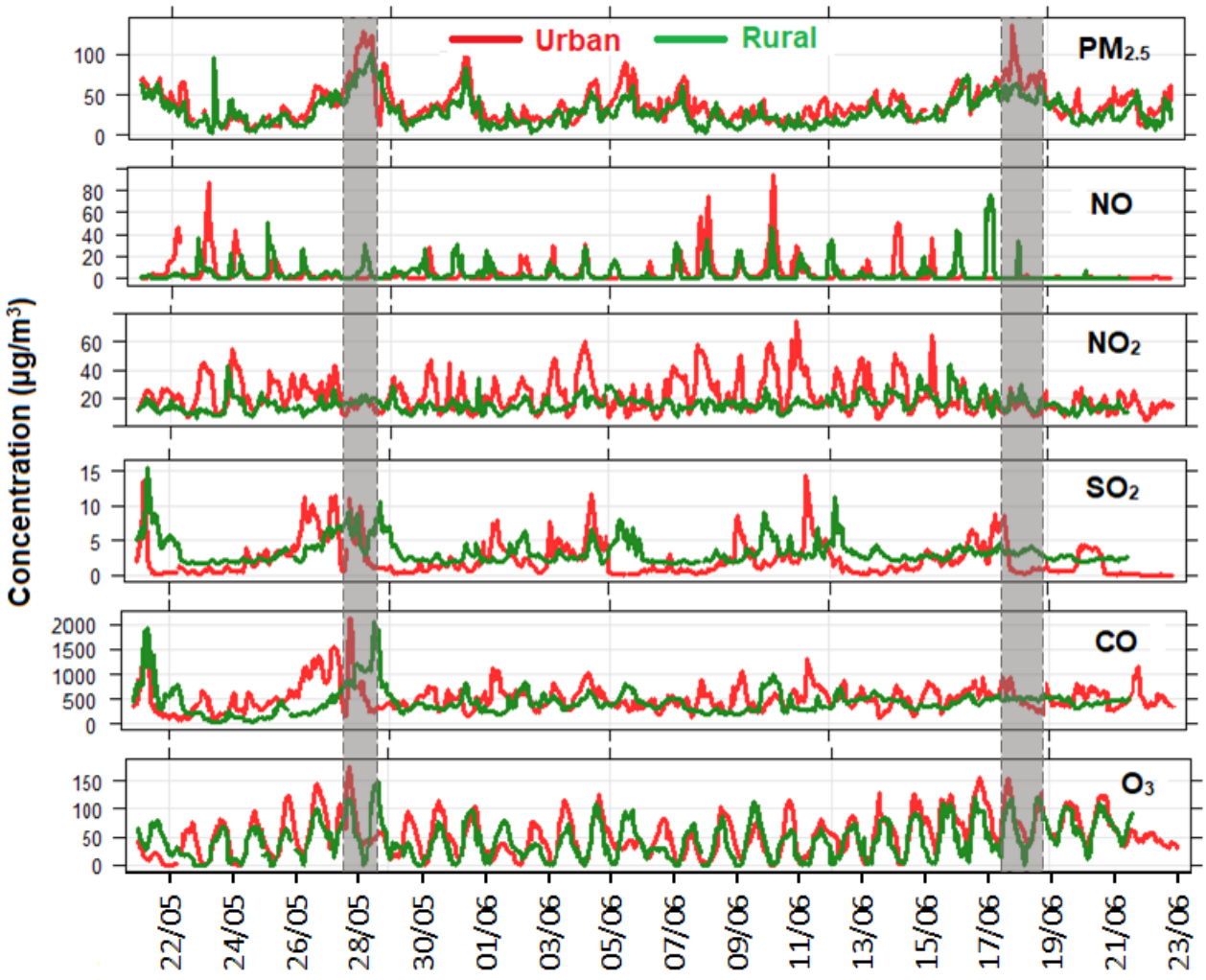


Figure 7: Time-series of air quality variables at the urban and rural sites during the summer campaign. Two minor haze events are indicated (shading).

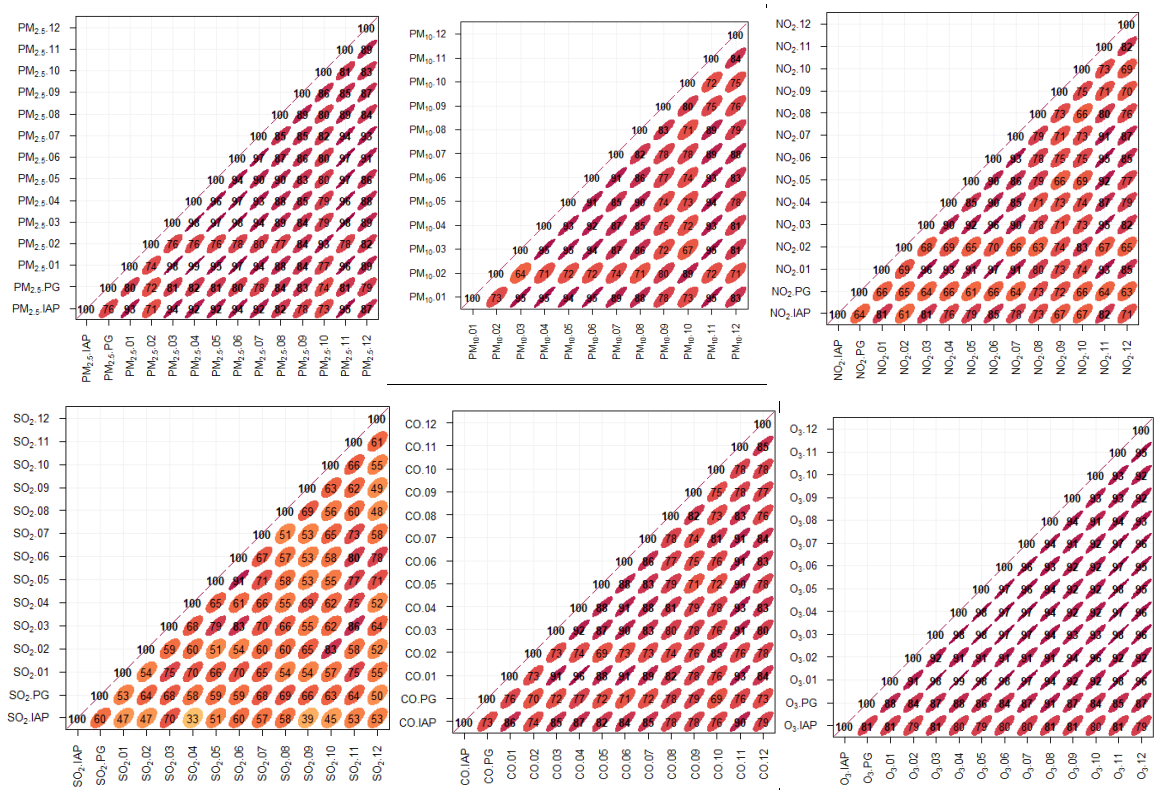
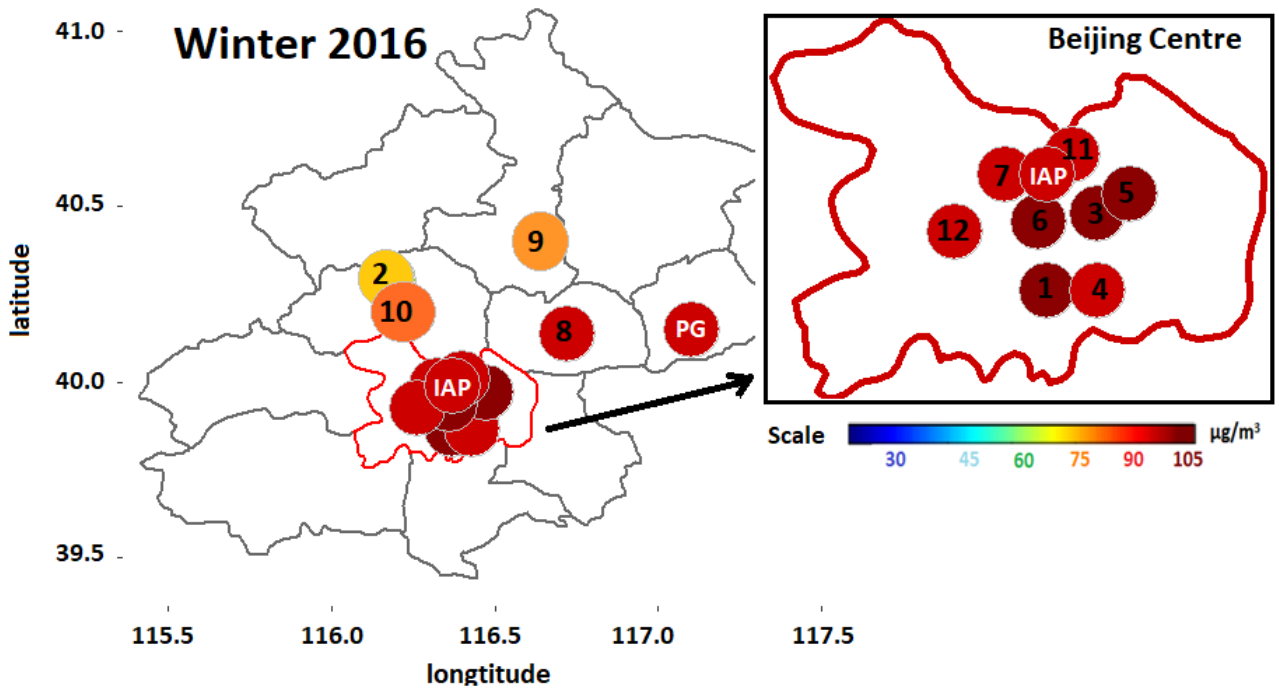


Figure 8: Correlations between the air quality at IAP, PQ and 12 monitoring station around Beijing. Stations G1-G12 (Figure 2) are labelled 01-12, PG = Pinggu.



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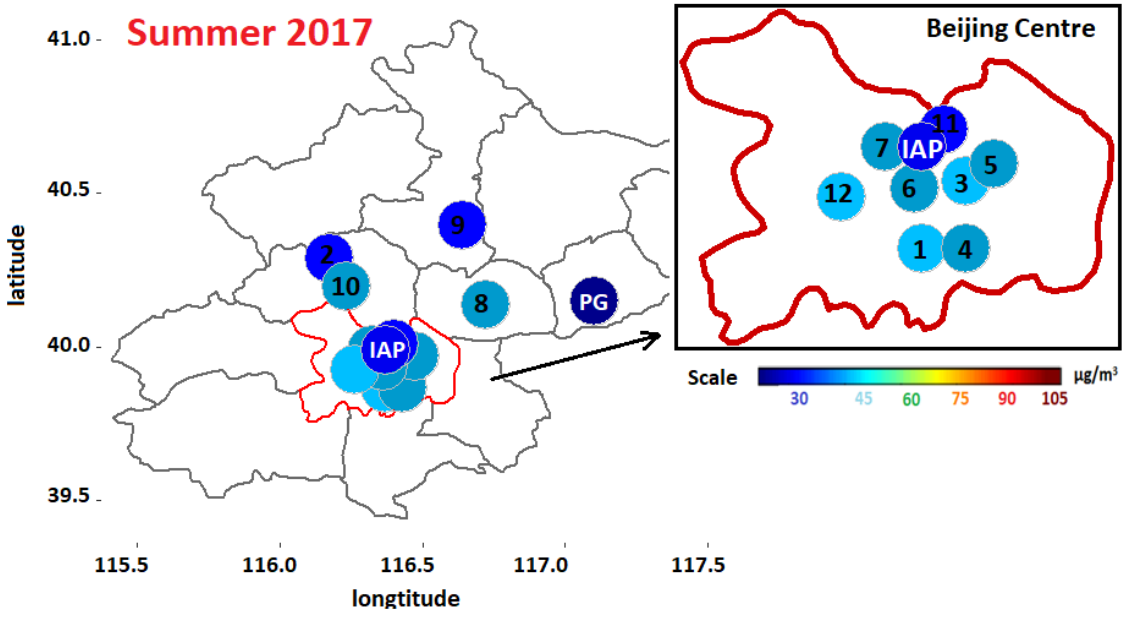

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1509 **Figure 9:** Spatial distribution of hourly mean concentration of $PM_{2.5}$ in Beijing during two

1510 sampling campaigns.

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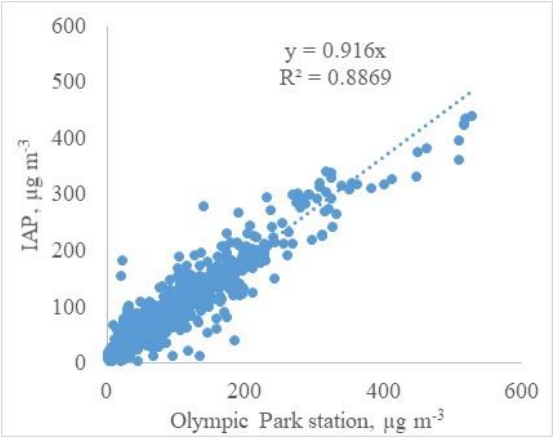


Figure 10: Hourly $PM_{2.5}$ at IAP (roof of a two storied building) and the neighbouring Olympic park national air quality monitoring station during the winter and summer intensive field campaigns.

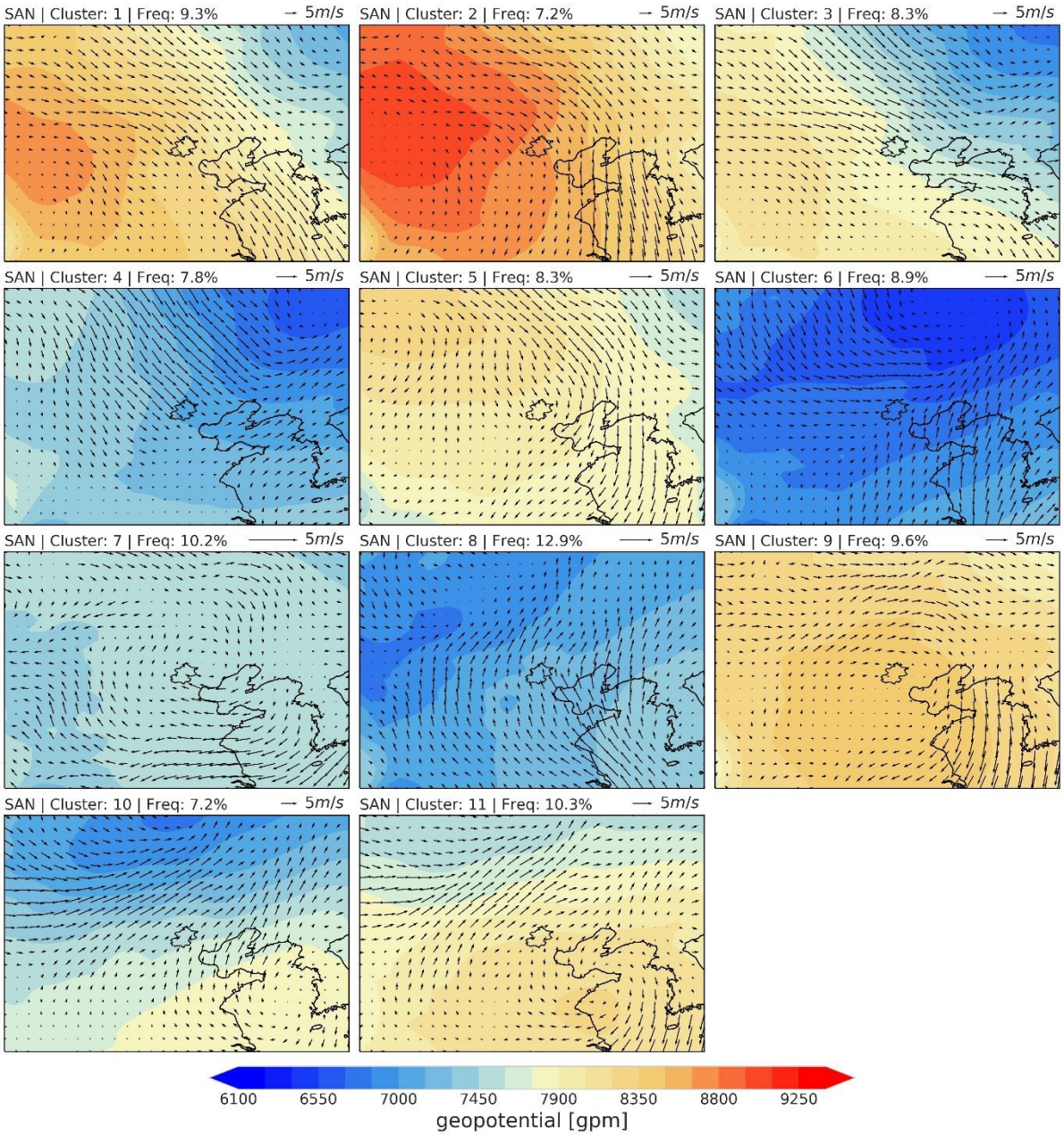


Figure 11: ERA-Interim (1988-2017) average 925 hPa geopotential with 10 m horizontal wind vector for 11 circulation types classified for Beijing (municipal boundary thin solid line) surroundings (103-129° E, 31 - 49° N) determined with the SANDRA method (COST733 class software). Frequency of occurrence is given in cluster caption. For discussion of conditions associated with each CT see section 6.1.

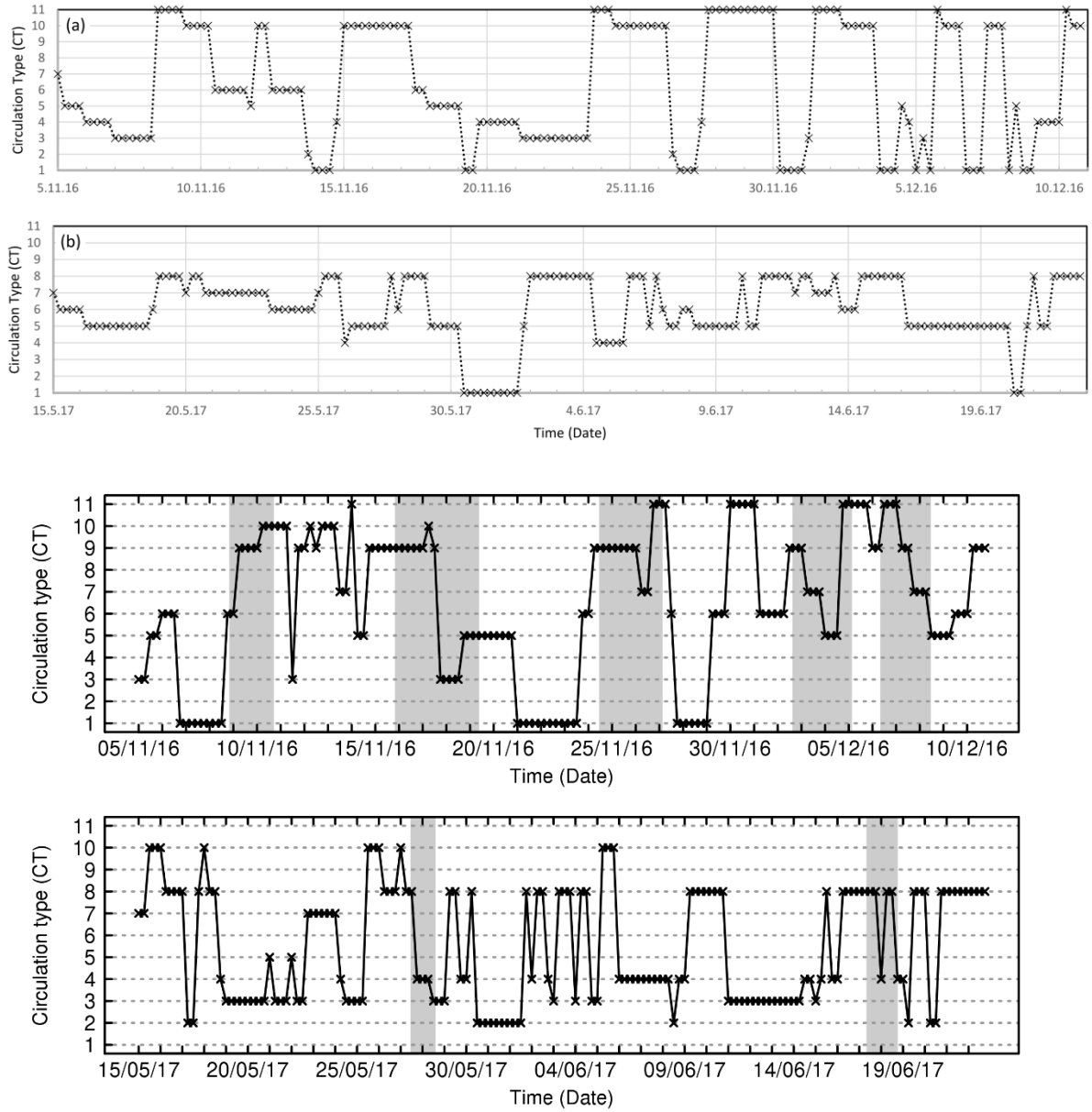


Figure 12: Time series of circulation types (CTs) during the two field campaigns: (a) winter and (b) summer. The 11 CTs are shown in Figure 11. See text for more description. Shading shows the pollution events identified in Section 5.

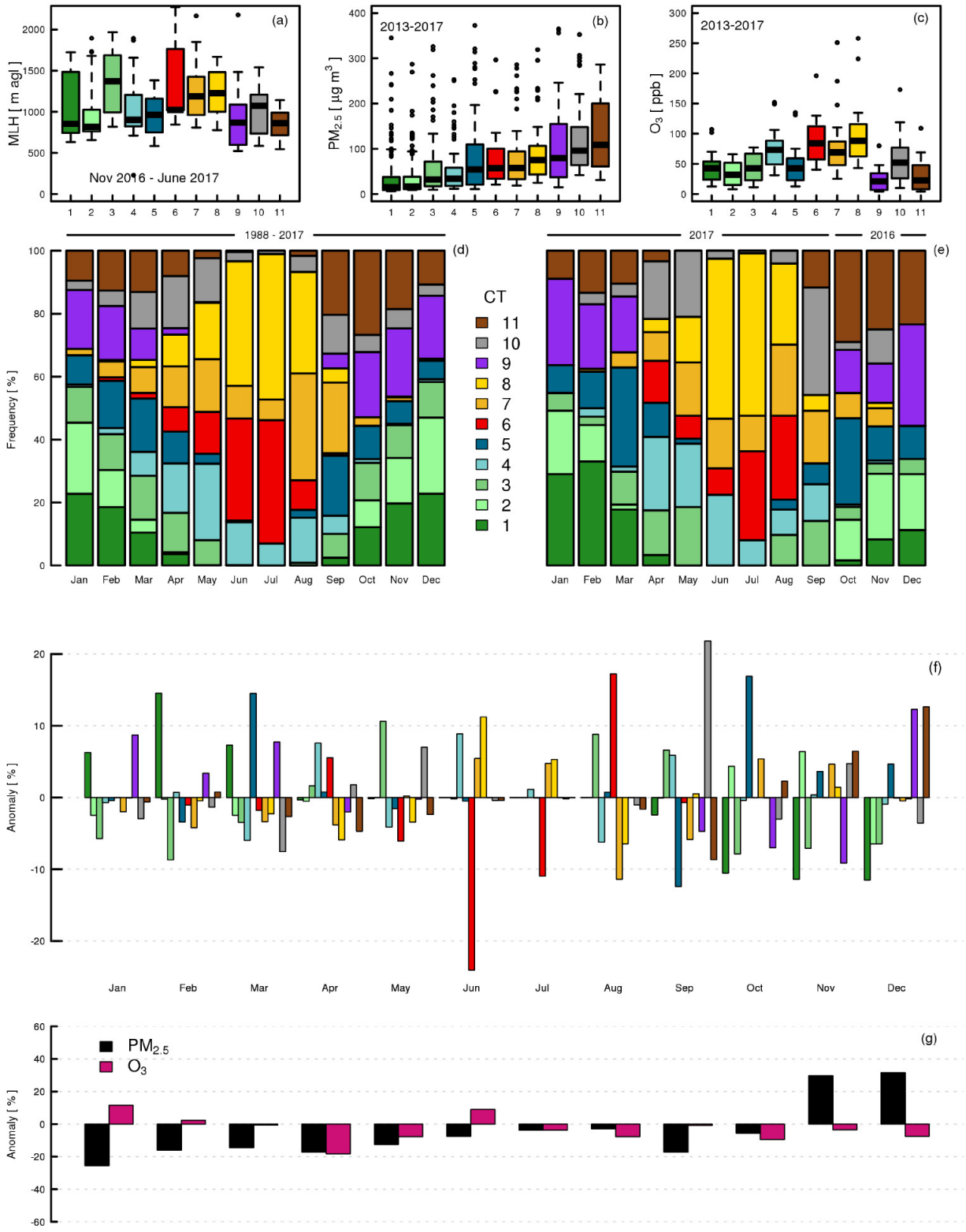


Figure 13: Analysis by circulation type (CT; Sect. 0) of: (a) daily maximum mixed layer height (MLH) determined from ALC observations at IAP between November 2016 – June 2017 (analysis method, Kotthaus and Grimmond, 2018b); concentration of (b) $PM_{2.5}$ and (c) O_3 at at the Olympic Park (i.e. Aotizhongxin) in 2013-2017 from the national air quality network; occurrence of CTs in

(d) 1988-2017 and (e) Oct 2016 – Sept 2017; (f) anomaly of CT frequency during Oct 2016 – Sept 2017 compared to 30 y climatology; and (g) anomaly of $PM_{2.5}$ and O_3 during Oct 2016 – Sept 2017 compared to 5 y (2013-2017) average (same data as in b, c).

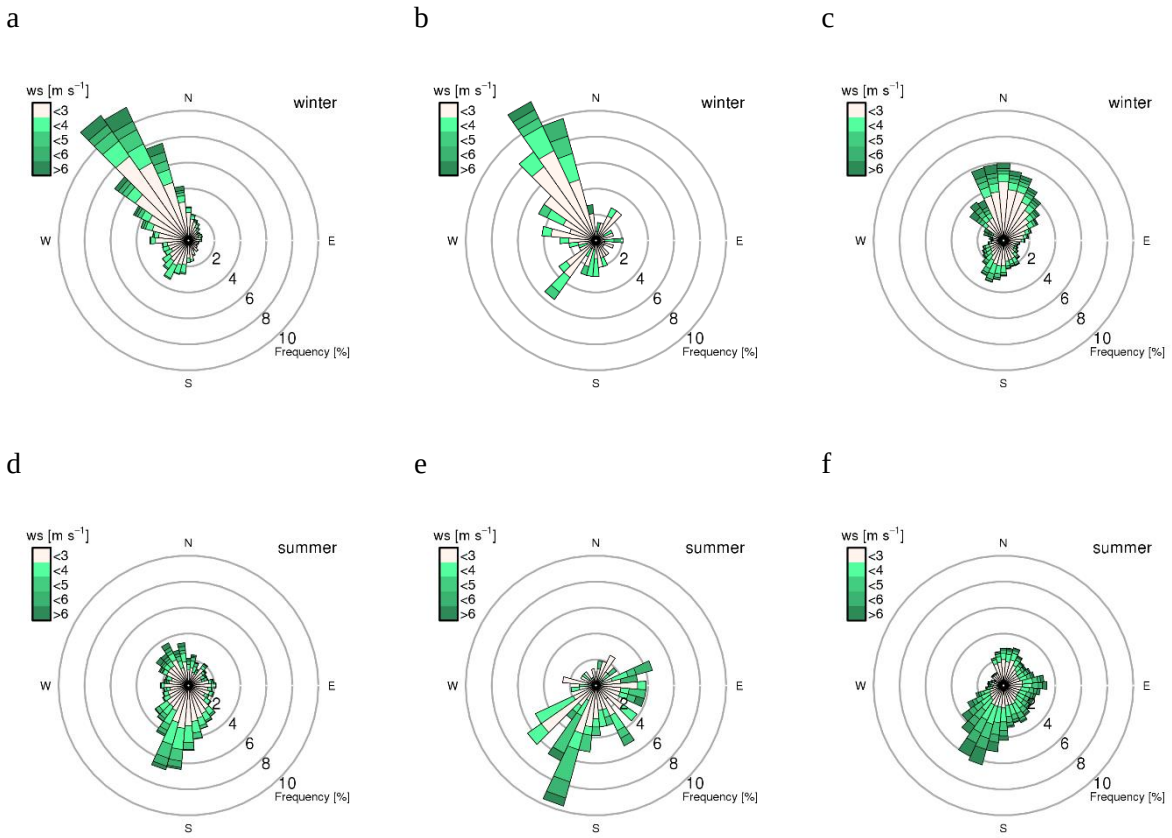
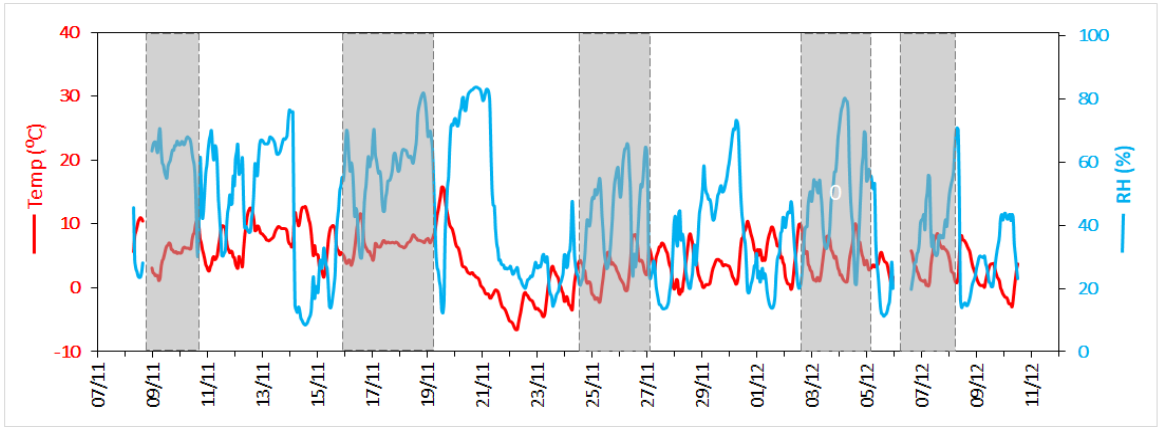


Figure 14: Beijing wind roses: (a, b, d, e) ERA-Interim 10 m horizontal wind (40° N, 116.5° E) and (c, f) sonic anemometer (Table 1) at IAP 320 m agl for (a) 5 November – 10 December in 1988-2017, (d) 15 May – 22 June in 1988-2017, (b, c) 5 November – 10 December 2016, and (e, f) 15 May – 22 June 2017.



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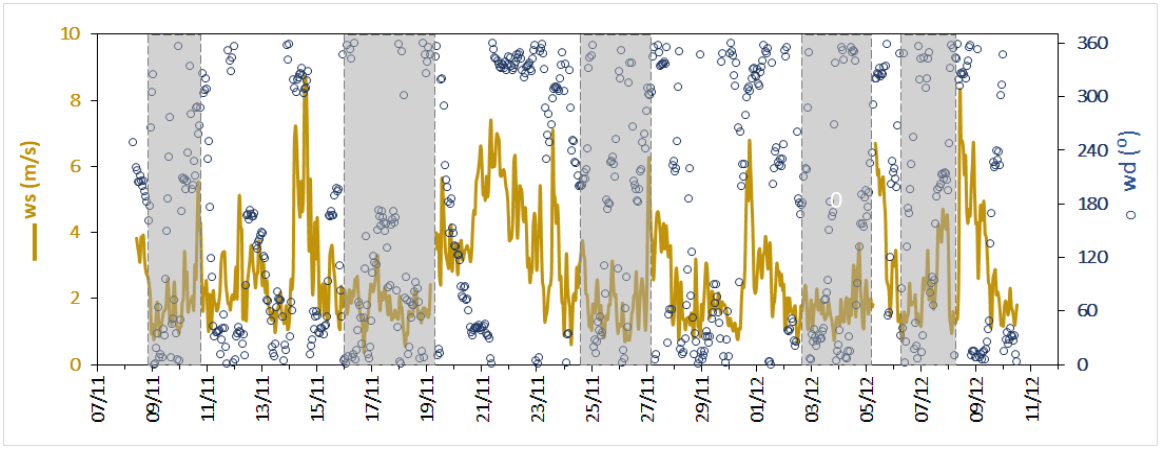

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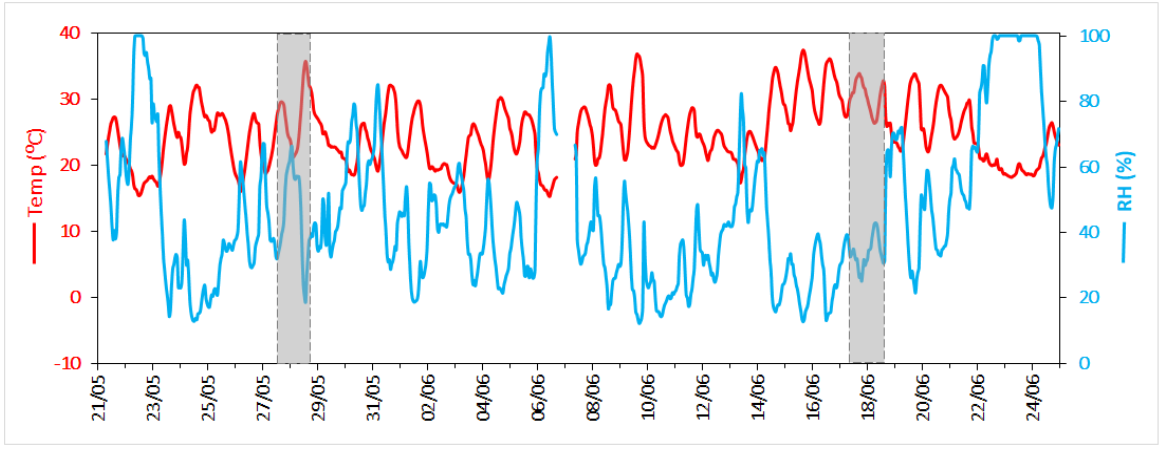

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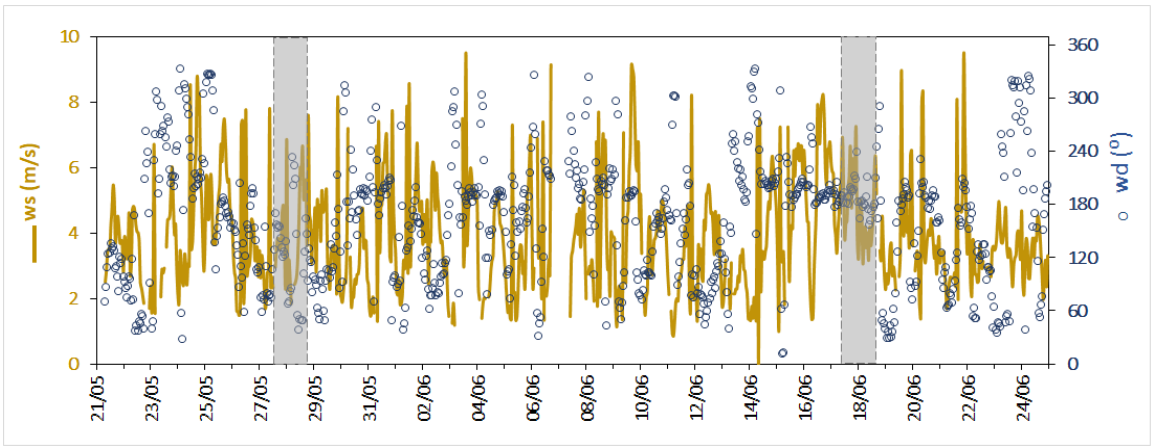

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Figure 15: Hourly meteorological variables measured at 120 m during the (a) winter and (b) summer campaigns. The shaded areas highlighted the haze periods (Table 3, Figures 2 and 7).

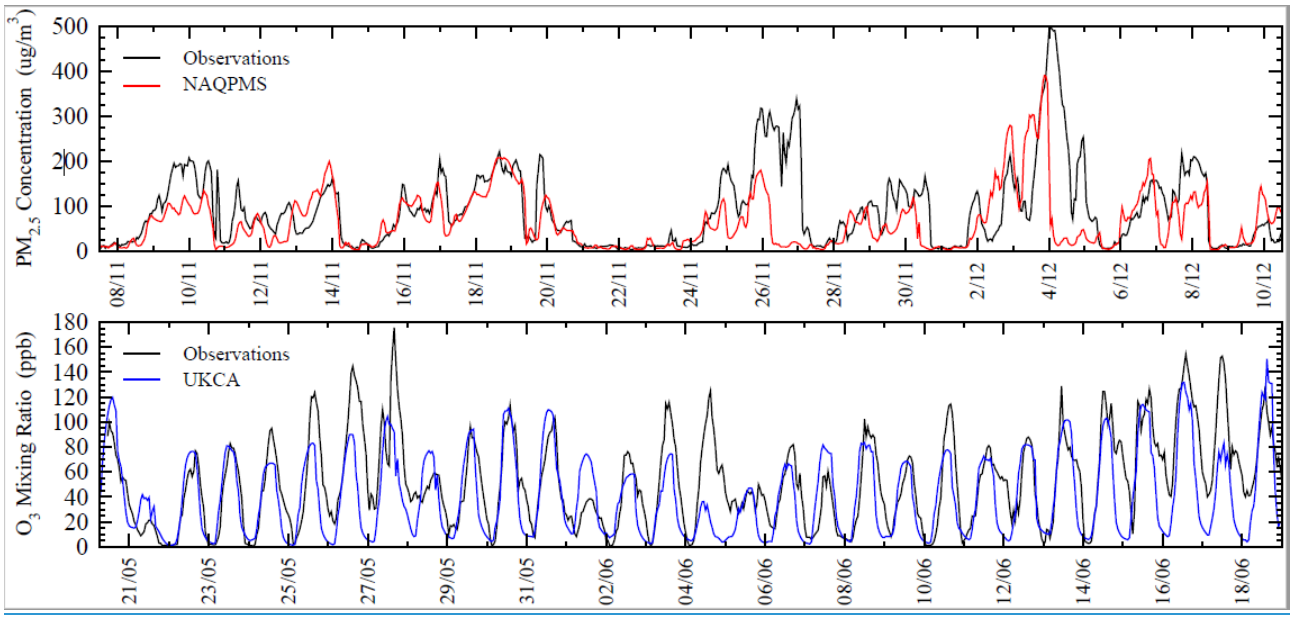

Figure 16: Comparison of observed and modelled pollutant concentrations showing (a) $PM_{2.5}$ concentrations during the winter campaign compared with NAQPMS simulations, and (b) O_3 mixing ratios in summer compared with UKCA simulations.

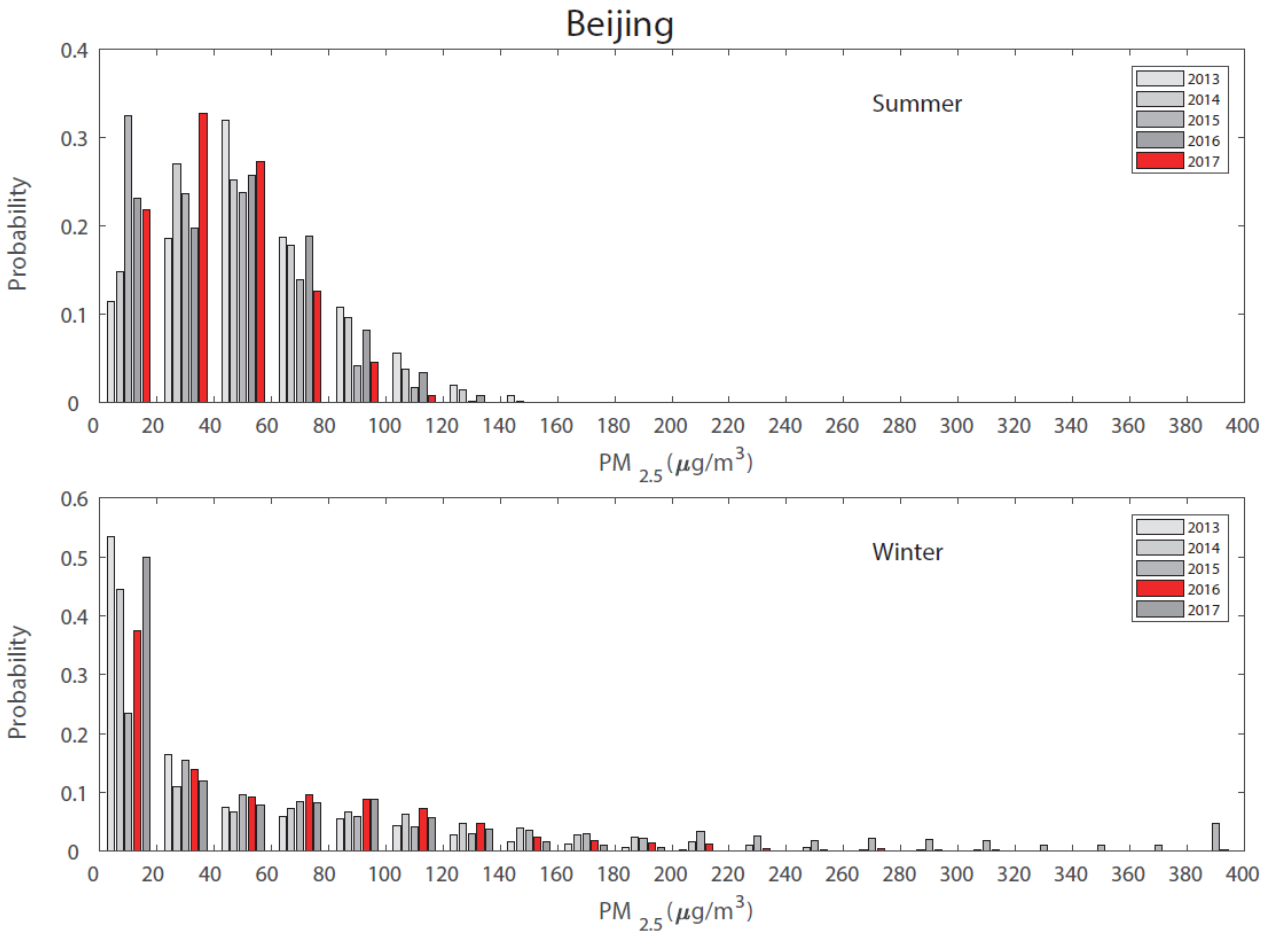

Figure ~~16~~17: Frequency distribution of $PM_{2.5}$ in Beijing over the summer (top) and winter (bottom) campaign periods from the NAQPMS model compared with those from the same periods over the past five years under the same emission conditions