Seasonal characteristics of forecasting uncertainties in surface PM$_{2.5}$ concentration associated with leading-time over the Beijing-Tianjin-Hebei region

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ABSTRACT

Forecasting uncertainties in meteorological fields have long been recognized as the main limitation on the accuracy and predictability of air quality forecasts. However, the impacts of meteorological forecasting uncertainties on air quality forecasts in different seasons are still not well known. In this study, a series of forecasts with different leading-time for January, April, July, and October of 2018 are conducted over the Beijing-Tianjin-Hebei (BTH) region, and the impacts of meteorological forecasting uncertainties on surface PM$_{2.5}$ concentration forecasts with each leading-time are investigated. With increasing leading-time, the forecasted PM$_{2.5}$ concentrations change significantly and have obvious seasonal variation. In general, the forecasting uncertainties in monthly mean surface PM$_{2.5}$ concentrations in the BTH region due to leading-time are the largest (80%) in spring, followed by autumn (~50%), summer (~40%), and winter (20%). In winter, the forecasting uncertainties in total surface PM$_{2.5}$ mass due to leading-time are mainly due to the uncertainties in PBL heights and hence the PBL mixing of anthropogenic primary particles. In spring, the forecasting uncertainties are mainly from the impacts of leading-time on the northwesterly, thereby further enhancing the condensation production of anthropogenic...
secondary particles by long-range transported natural dust. In summer, the forecasting uncertainties result mainly from the decrease of dry and wet deposition rates, which are associated with the reduction of near-surface wind speed and precipitation rate. In autumn, the forecasting uncertainties are mainly from the change in the transport of remote natural dust and anthropogenic particles, which is associated with the change in large-scale circulation.

**Key words:** PM$_{2.5}$; Forecasting uncertainties; Leading-time; Meteorological fields; The Beijing-Tianjin-Hebei region

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**Article Highlights:**

- Forecasting uncertainties of PM$_{2.5}$ due to leading-time are large.
- Forecasting uncertainties of PM$_{2.5}$ are largest (smallest) in spring (winter).
- Improving the forecast accuracy of key meteorological fields needs more effort.

**1. Introduction**

As one of China's most densely populated and urbanized megalopolis clusters, the Beijing-Tianjin-Hebei (BTH) region has suffered from serious air pollution in recent years, especially heavy PM$_{2.5}$ pollution. Ambient PM$_{2.5}$ is a great threat to human health (e.g., Chen and Hoek, 2020; Ho et al., 2018; Yang et al., 2019; Yue et al., 2021). In addition, these atmospheric particles can also interact with radiation, and alter cloud microphysics and precipitation, thereby affecting the radiative energy balance of the Earth’s system (e.g.,
Since 2013, there have been many heavy air pollution events covering most areas of China where the 24 h average concentrations of PM\textsubscript{2.5} exceed the threshold of ‘severe pollution’ identified by China’s ambient air quality standards (GB3095-2012). To control air pollution, the Chinese government has issued many policies and measures, including the promotion of energy conservation and emission reduction (e.g., Chen et al., 2020; Chen et al., 2019; Fang et al., 2019; Ji et al., 2019; Kanaya et al., 2020; Qu et al., 2020; Zheng et al., 2018). However, the mechanisms of the generation and evolution of PM\textsubscript{2.5} pollution remain unclear. Moreover, global climate change and the frequent occurrence of extreme weather also influence PM\textsubscript{2.5} evolution and the interactions of aerosol and meteorological processes, making it more challenging to effectively control PM\textsubscript{2.5} pollution in China.

Air quality forecasting is critical for administrators to develop efficient and effective strategies for air pollution control. The forecast of air pollution episodes a few days in advance can help the government take short-term actions and issue timely alerts to reduce the exposure of the population and prevent potential pollution (e.g., Honore et al., 2008; Liu et al., 2018; Menut et al., 2015; Petersen et al., 2019). Generally, there are two kinds of methods to forecast air quality, i.e., statistical models and numerical models. Although statistical methods can predict air pollution episodes well, they have limited capability to reveal the kinetic and chemical mechanisms of air pollution (Agarwal et al., 2020; Chae et al., 2021; Gupta and Christopher, 2009; Lee et al., 2011; Liu et al., 2005; Seng et al., 2021; Xiao et al., 2020). In contrast, numerical models take meteorology and chemistry processes into account, enabling the analysis and understanding of chemical and physical processes affecting the formation and evolution of air pollution. This method has been widely used...
for air quality forecasting and studying the temporal and spatial variations in air pollutants (e.g., Liu et al., 2018; Ni et al., 2018; Bao et al., 2019; Khan and Kumar, 2019; Kumar et al., 2020; Petersen et al., 2019; Spiridonov et al., 2021; Spiridonov et al., 2019). However, as of now, accurate forecasting of air pollution episodes remains a challenge.

The accurate forecasting of air quality with numerical models is greatly hindered by the uncertainties in model input fields (e.g., emissions and initial conditions) and the incomplete or insufficient parameterization of the relevant atmospheric chemical processes. In addition, meteorological forecast uncertainties also largely affect air quality forecast accuracy since they can directly affect processes such as transport, turbulent mixing, and dry deposition (e.g., Chatani and Sharma, 2018; Dennis et al., 2010; Gilliam et al., 2015; Hu et al., 2019a; McNider and Pour-Biazar, 2020; Podrascanin, 2019; Rao et al., 2020; Willmott et al., 1985). The impacts of meteorological forecast uncertainties on air quality forecasts have been widely studied (e.g., Bei et al., 2014; Delle Monache et al., 2006; Garcia-Menendez et al., 2013; Liu and Wang, 2020; Thomas et al., 2019; Zhang et al., 2007; Zhao et al., 2019). Zhao et al. (2019) investigated the influence of planetary boundary layer (PBL) and surface layer parameterizations on surface ozone concentrations in the BTH region. They found that the choices of PBL and surface layer schemes change the simulated surface ozone concentrations by an over 20% difference in ozone dry deposition fluxes. Liu and Wang, (2020) examined the contribution of the variations in meteorological conditions and anthropogenic emissions to the summer ozone concentration from 2013 to 2017 in China. They found that in some areas of China, changes in meteorological conditions may have a more significant effect on ozone than anthropogenic emissions, with changes in wind fields making an important contribution to
the increase in surface ozone. Garcia-Menendez et al., (2013) investigated the sensitivity
of forecasted PM$_{2.5}$ concentrations to uncertain wind fields and found that the uncertainties
of forecasted PM$_{2.5}$ concentrations might be determined by the uncertainties of wind fields
rather than emissions. In addition, they found that the NME (normalized mean error) of the
forecasted PM$_{2.5}$ concentrations caused by the uncertainties in wind fields could reach
100%.

Many studies have proven that the numerical forecast uncertainties of
meteorological fields generally increase with increasing leading-time (e.g., Baumgart et
al., 2019; Judt, 2018; Parsons et al., 2019; Selz, 2019; Zhang et al., 2019), which largely
limits the forecast accuracy of air quality. Therefore, it is also important to examine the
impacts of leading-time on air quality forecasts to explore the factors affecting the forecast
accuracy of air quality, which has been investigated by some previous studies (e.g., de
Wildt et al., 2011; Emili et al., 2016; Gvozdikova and Muller, 2021; Honore et al., 2008;
Liu et al., 2018; Menut et al., 2015; Neal et al., 2014; Spiridonov et al., 2019; Thomas et
al., 2019; Zhang et al., 2007; Zhang et al., 2017). Honore et al., (2008) conducted forecast
experiments over Europe from 2004 to 2006 with leading-times from 24 to 72 hours. By
evaluating the surface ozone concentrations with observations, they found that 6%, 15%,
and 25% of the ozone daily maxima forecast uncertainties of the 24, 48, and 72-hour
forecasts are due to meteorological forecast uncertainties at some rural stations. de Wildt
et al., (2011) examined the ability of the Lotos-Euros model to forecast PM$_{10}$
concentrations of 2009 for a leading-time of 6 days in the Netherlands. The correlation
between the forecast and observations is approximately 0.66-0.77 with a leading-time of
less than 3 days and then gradually deteriorates to 0.54 for the 6 days forecast. Zhang et
al., (2017) evaluated the predictability of hourly and daily PM$_{2.5}$ of WRF-Chem in downtown Beijing for November–December 2015 with a leading-time range of 10 days. They found that the root mean square error (RMSE) of the forecasted hourly (daily) mean PM$_{2.5}$ increased significantly with increasing leading-time, rising from 103.1 (85.8) for the 1 day forecast to 150.7 (130.0) for the 10 days forecast.

Some previous studies have reported the impacts of leading-time on air quality forecast results. Few studies focused on the impact of leading-time on air quality forecast in China on a seasonal scale and investigated the mechanisms driving the air quality forecast uncertainties caused by increasing leading-time for different seasons. It is not well known which processes are the keys that result in forecasting uncertainties with increasing leading-time. Therefore, in this study, a few air quality forecast experiments are conducted with different leading-times for four seasons over China. The analysis aims to explore the impacts of meteorological forecast uncertainties with increasing leading-time on the forecasting uncertainties of surface PM$_{2.5}$ concentrations in different seasons, and the underlying mechanisms are also investigated. This study focuses on the BTH region, including Beijing, Tianjin and Hebei Province, as shown in Figure 1. The BTH region is located on the coast of the Bohai Sea, with the Taihang Mountains to the west, the Yanshan Mountains to the north, and the Yimeng Mountains to the southeast. With a large amount of heavy industry and seasonal heating demand, the BTH region has become the most polluted region by anthropogenic emissions in China and typically represents the characteristics of air pollution over northern China. In addition, the BTH region has a typical temperate and continental monsoon climate with distinct seasons, featuring hot and humid in summer and cold and dry in winter.
The rest of the paper is organized as follows. The details of the model used, the experimental design, and the analysis method of the impacts of meteorological forecast uncertainties are presented in Section 2. The quantitative analysis of the impacts of meteorology forecast uncertainties and the investigation of the underlying mechanisms are discussed in Section 3. The summary and discussion can be found in Sections 4 and 5, respectively.

2. Methodology

2.1 WRF-Chem

In this study, the USTC version of WRF-Chem with new functions including radiation forcing diagnosis of aerosol species and improved PBL mixing of aerosols is used (Du et al., 2020; Hu et al., 2019b; Zhang et al., 2021; Zhao et al., 2013a; Zhao et al., 2014; Zhao et al., 2016; Zhao et al., 2013b). In particular, to understand the mechanisms by which
uncertainties in meteorological forecasts affect the surface PM$_{2.5}$ concentrations forecasts, this version is updated with a detailed diagnosis of the contributions of each crucial process to surface PM$_{2.5}$ concentrations (Du et al., 2020). The WRF-Chem configurations of this study are similar to that of Du et al., (2020). Briefly, the CBM-Z (Zaveri and Peters, 1999) and the MOSAIC (Zaveri et al., 2008) are used for aerosol chemistry. The size distributions of aerosol particles are characterized by eight bins following the method of Fast et al., (2006). The description of dry depositions of aerosol mass and number follows the method of Binkowski and Shankar, (1995). The wet deposition of aerosols follows the method of Easter et al., (2004) and Chapman et al., (2009). The YSU PBL scheme (Hong et al., 2006) is used, and the PBL mixing coefficient within the boundary layer is modified following Du et al. (2020) by setting the lower limit of the PBL mixing coefficient from the default value in WRF-Chem to 5 m$^2$s$^{-1}$, and is applied in the mixing of gas and aerosol pollutions. The Morrison two-moment scheme is used for cloud physics (Morrison et al., 2009), while the RRTMG (Iacono et al., 2000; Mlawer et al., 1997; Zhao et al., 2011) is used for radiation. The description of aerosol optical properties follows the method of Fast et al., (2006) and Barnard et al., (2010). The description of activation and resuspension of aerosols in aerosol-cloud interactions follows the methods of Gustafson et al., (2007).

**2.2 Experimental design**

In this study, WRF-Chem is conducted with two nested domains (one-way nesting) in four months of 2018 (i.e., January, April, July, and October) as representatives of four seasons. The outer domain covers a quasi-global region with 360×145 grid cells (180°W–180°E, 67.5°S–77.5°N) at a 1°×1° horizontal resolution (not shown), while the inner domain covers East China with 147×196 grid cells (107.1°E–127.9°E, 30°N–53°N).
20.1°N~47.9°N) at a horizontal resolution of 0.14°×0.14°, as shown in Figure 1. Although
the analysis in this study focuses on the BTH region, this inner domain covers the entire
region of East China, including almost all densely populated regions of China, to consider
the impacts of transport. The preliminary analysis over other regions, such as the Yangtze
River Delta (YRD) region and the Pearl River Delta (PRD) region, shows quite different
results and mechanisms compared to those over the BTH region and therefore will be
presented in subsequent studies in the future. In addition, since the surface PM$_{2.5}$
concentrations are relatively low in the mountainous northwestern areas of the BTH region
(to be discussed in Section 3) and the populated urban areas are mainly distributed in the
central and southern areas of the BTH region, the analysis of this study will focus on the
central and southern plain areas of the BTH region (Fig. S1).

The forecast experiments are conducted with the two nested domains for four
months of 2018. Each forecast experiment starts at 00 UTC each day of the month and runs
for 144 hours. In this way, the composite forecasting results can be obtained for each month
with the leading-time (the time span of the forecast) from 24 hours to 144 hours, and the
impacts of the leading-time induced by meteorological forecasting uncertainties can be
investigated. Specifically, the composite of the average forecasting results between 24
hours and 48 hours is referred to hereafter as the 2-Day forecast, and analogously, the 3-
Day, 4-Day, 5-Day, 6-Day, and 7-Day forecasts are designated for analysis. For each
forecast run, the chemical initial fields and lateral conditions of the inner domain are
obtained from the outer quasi-global domain, and the meteorological initial conditions of
both domains are obtained from the ERA-interim reanalysis data. Meanwhile, the chemical
initial fields and lateral conditions of the forecast in the outer quasi-global domain are
obtained by a quasi-global simulation with the same settings (Table 1), except the four-
dimensional data assimilation (FDDA) is applied (Stauffer and Seaman, 1990; Seaman et
al., 1995). As the outer domain covers almost the entire globe, although the experiments
are driven by the reanalysis at the meridional lateral boundaries, the simulation results can
be treated as forecast results. Please note, a few test experiments are also conducted with
the ERA5 reanalysis as initial and boundary conditions. A negligible difference is found
between the results driven by different reanalysis (not shown), which indicates that the
results in this study do not depend on the reanalysis used.

The configuration of emissions in this study is similar to that used by Du et al., (2020).
For both domains, the monthly emission inventory of Hemispheric Transport of Air
Pollution version 2 (HTAPv2) of 2010 at 0.1°×0.1° horizontal resolution (Janssens-
Maenhout et al., 2015) is used with the emissions over China replaced by the 0.1°×0.1°
Multi-resolution Emission Inventory for China (MEIC) of 2015 (Li et al., 2017a; Li et al.,
2017b). The biomass burning emissions are derived from the hourly Fire Inventory from
NCAR (FINN) at 1 km resolution (Wiedinmyer et al., 2011), which follows the injection
heights proposed by Dentener et al., (2006) in the Aerosol Comparison between
Observations and Models (AeroCom) and the diurnal variation provided by WRAP (2005).
The biogenic emissions are simulated by MEGAN v2.0 following Zhang et al., (2021). In
addition, the GOCART dust emission scheme (Ginoux et al., 2001) is used to calculate the
vertical dust flux, and the size distributions of dust particles emitted into the atmosphere
are characterized in the MOSAIC aerosol size bins (Kok, 2011). Besides, the sea-salt
emission follows Zhao et al., (2013b).
2.3 Diagnosis of impacts of meteorological forecast uncertainties

Normally, the forecasting uncertainties can be assessed by comparing the forecasting results and the reanalysis or observations. However, in addition to the impacts of forecast uncertainties of meteorological fields, air quality forecasting uncertainties may result from many other factors, such as uncertainties of emissions and deficiencies in physical and chemical parameterizations, and the uncertainties due to these factors may cancel each other out. Therefore, the impacts of meteorological forecast uncertainties caused by leading-time cannot be isolated through comparison with reanalysis or observations. In this study, the 2-Day forecast is treated as the benchmark, and the impacts of meteorological forecast uncertainties caused by leading-time on the forecast uncertainties of surface PM$_{2.5}$ concentration are analyzed by the difference in forecasted PM$_{2.5}$ concentrations between the 3-Day to 7-Day forecasts and the 2-Day forecast. Some key meteorological fields from the 2-Day forecast are evaluated with the ERA5 reanalysis data. The forecast generally captures the spatial features of the geopotential heights and winds at 850 hPa for different seasons (Fig. S2 in the supporting material). The forecast bias of geopotential heights is less than 50 m in east China in winter and within 10 m in other seasons, while the bias of winds is less than 5 m s$^{-1}$ in east China in all seasons. In addition, the forecast slightly overestimates (underestimates) the temperature at 2 m in central and eastern China (in the northern and southern coasts) with biases less than 3 K (Fig. S3 in the supporting material). In general, the meteorological fields from the 2-Day forecast are reliable. Therefore, by comparing the differences in PM$_{2.5}$ concentrations between the 3-Day to 7-Day forecasts and the 2-Day forecast, the impacts of meteorological forecast uncertainties due to leading-time can be quantified.
3. Results

3.1 Forecasting uncertainties of surface PM$_{2.5}$ concentrations

First, the spatial distributions of surface PM$_{2.5}$ concentrations from the 2-Day to 7-Day forecasts are shown in Figure 2. The surface PM$_{2.5}$ concentrations from the 2-Day forecast are higher in the central and southern areas in the BTH region throughout the seasons, especially in the plain areas. The PM$_{2.5}$ concentrations over the mountainous northwestern areas from the 2-Day forecast are less than 30 ug m$^{-3}$ throughout the year, and therefore, this area will not be the focus of the study. The highest PM$_{2.5}$ concentrations throughout the year occur in winter, with concentrations up to 70 ug m$^{-3}$. In spring, surface PM$_{2.5}$ concentrations are also relatively higher over the Northwest, which is due to long-range transported dust prevailing in this season (discussed more below). In summer, the surface PM$_{2.5}$ shows a similar spatial distribution to that in winter, with a lower concentration of approximately 20 to 50 ug m$^{-3}$. However, the PM$_{2.5}$ concentrations are below 40 ug m$^{-3}$ in the entire area in autumn. With increasing leading-time, the spatial distributions of forecasted PM$_{2.5}$ concentrations change significantly and have obvious seasonal differences. The forecasted PM$_{2.5}$ concentrations continue to increase with increasing leading-time over the BTH region throughout the four seasons, except that the concentrations may decrease in winter and autumn.
Figure 2. Spatial distribution of surface PM$_{2.5}$ concentrations from the 2-Day to 7-Day forecasts in January, April, July, and October of 2018.

Figure 3 shows the regional surface PM$_{2.5}$ concentrations averaged over the plain areas of BTH (as shown in Fig. S1) for four seasons from the forecasts with different leading-time. In winter, the surface PM$_{2.5}$ concentration from the 2-Day forecast is approximately 55 µg m$^{-3}$, and it increases with the increasing leading-time, peaking at 65 µg m$^{-3}$ in the 4-Day forecast. It then decreases to approximately 53 µg m$^{-3}$ in the 7-Day forecast. This indicates that the forecasting uncertainties of monthly mean surface PM$_{2.5}$ concentrations in the BTH region in winter can reach 20% solely due to the forecasting meteorological
uncertainties with a 4-Day or longer leading-time. In spring, with the increasing leading-time, the forecasted PM$_{2.5}$ concentrations increase from 32 ug m$^{-3}$ in the 2-Day forecast to 58 ug m$^{-3}$ in the 6-Day forecast. The impacts of leading-time on the forecasting uncertainties of surface PM$_{2.5}$ concentrations are much larger (~80%) in spring than in winter (~20%) over the BTH region. In summer, the forecasted surface PM$_{2.5}$ concentrations generally increase with the leading-time from 27 ug m$^{-3}$ in the 2-Day forecast to 36 ug m$^{-3}$ in the 7-Day forecast, reflecting a forecasting uncertainty of ~40%.

In autumn, the impact of leading-time on forecasted surface PM$_{2.5}$ concentrations is similar to that in summer in the 2-Day to 5-Day forecasts (~50%) but is different in the 6-Day and 7-Day forecasts. Overall, the forecasting uncertainties in surface PM$_{2.5}$ concentrations are the smallest in winter among the four seasons and can reach more than 80% solely due to the forecasting meteorological uncertainties with leading-time of 7 days or even shorter.

**Figure 3.** Monthly mean surface PM$_{2.5}$ concentrations over the BTH region in January, April, July, and October of 2018 from the 2-Day to 7-Day forecasts.
3.2 Factors driving the forecasting uncertainties

To further understand the seasonal characteristics of the forecasting uncertainties of surface PM$_{2.5}$ concentrations due to the increasing leading-time, Figure 4 shows the regional surface concentrations of PM$_{2.5}$ compositions averaged over the plain areas of BTH for four seasons from the forecasts with different leading-times. From the 2-Day forecast, in winter, anthropogenic primary particles dominate the total surface PM$_{2.5}$, i.e., the masses of OC, BC, and OIN occupy ~70% of the total PM$_{2.5}$ mass. The masses of anthropogenic secondary particles (i.e., nitrate, sulfate, ammonia) occupy ~24%. These six species show similar trends with increasing leading-time, which is also consistent with the pattern of total PM$_{2.5}$ mass (Fig. 3). The change in surface PM$_{2.5}$ with leading-time corresponds well to the change in PBL height. Figure 5 shows the regional averaged PBL heights over the plain areas of BTH for four seasons from the forecasts with different leading-times. It is obvious that the PBL height increases since the 4-Day leading-time in winter, which leads to a decrease in the forecasted surface anthropogenic primary and secondary particles and thus the total PM$_{2.5}$ mass. In addition, the dry deposition rate also increases with the leading-time (Fig. 6), particularly after the leading-time of 4-Day, which also leads to a decrease in the surface concentrations of anthropogenic particles with the leading-time. In WRF-Chem, the dry deposition velocity of particles is proportional to the near-surface wind with a positive relationship (e.g., Peters and Eiden, 1992; Wesely and Hicks, 1977). Therefore, this change in the dry deposition rate results from the increase of the near-surface wind speed due to the increasing leading-time (Fig. 7).
Figure 4. Monthly mean surface concentrations of PM$_{2.5}$ compositions over the BTH region in January, April, July, and October of 2018 from the 2-Day to 7-Day forecasts.
Figure 5. Monthly mean PBL heights over the BTH region in January, April, July, and October of 2018 from the 2-Day to 7-Day forecasts.

Figure 6. Same as Fig. 5, but for dry deposition velocity.
It is also noteworthy that the mass of natural dust particles increases significantly with leading-time from 1 ug m$^{-3}$ in the 2-Day forecast to 15 ug m$^{-3}$ in the 7-Day forecast, leading to the contribution from a negligible amount to almost 30%. Figure 8 shows the spatial distributions of wind fields at 700 hPa for the four seasons from the forecasts with different leading-times. Over the plain area of BTH, natural dust is mainly transported from the northwest. It is obvious that the northwesterly become stronger with increasing leading-time in winter, which leads to an increase in long-range transported dust.
Figure 8. Spatial distribution of wind circulations at 700 hPa from the 2-Day to 7-Day forecasts in January, April, July, and October of 2018.
In spring, the surface concentrations of anthropogenic primary particles are much lower than those in winter. In fact, they are much lower in the other three seasons than in winter. This is due to their much lower emissions in seasons other than winter, particularly for OC and BC (Fig. S8 in the supporting material). The masses of these three anthropogenic primary species occupy approximately 30% of the total surface PM$_{2.5}$ mass from the 2-D forecast. Natural dust and anthropogenic secondary particles both contribute one-third of the total surface PM$_{2.5}$ mass in the 2-D forecast. With the increasing leading-time, the masses of anthropogenic primary species increase slightly, corresponding to the small variation in PBL height, while the masses of natural dust and anthropogenic secondary particles increase more significantly. The total mass of anthropogenic secondary particles increases from 9 ug m$^{-3}$ in the 2-Day forecast to 15 ug m$^{-3}$ in the 7-Day forecast. The change of near-surface temperature is relatively small, corresponding to the increasing leading-time (Fig. 9), which should have a small impact on the production of secondary particles. This increase in anthropogenic secondary particles is likely due to condensation formation over the increase in the existing particle surface area from natural dust. The mass of natural dust increases significantly from 10 ug m$^{-3}$ in the 2-Day forecast to more than 20 ug m$^{-3}$ in the 7-Day forecast, which results from the stronger northwesterly with the increasing leading-time in spring (Fig. 8). The change in the dry deposition rate due to the increasing leading-time is also small. Therefore, the impacts of increasing leading-time on the forecasted surface PM$_{2.5}$ concentrations in spring are mainly through its impacts on large-scale circulation and thus on long-range transported dust, which can further induce changes in the production of anthropogenic secondary particles.
Figure 9. Spatial distribution of temperature at 2 m from the 2-Day to 7-Day forecasts in January, April, July, and October of 2018.

In summer, the total mass of surface PM$_{2.5}$ is the lowest among the four seasons (Fig. 3). Anthropogenic primary and secondary particles occupy approximately 40% and 60%, respectively, of the total surface PM$_{2.5}$ mass in the 2-Day forecast (Fig. 4). With the increasing leading-time, the slight increase in the total surface PM$_{2.5}$ mass mainly results from the increase in anthropogenic secondary particles. The increase in total mass in the 6-Day and 7-Day forecasts also comes partly from the slight increase in natural dust. In general, the PBL height varies slightly and should have a relatively small impact in summer (Fig. 5). This increase in the surface mass of particles may correspond to the decrease in the dry and wet deposition rates (Fig. 6), which results from the decrease in the near-surface wind speed and precipitation rate with the increasing leading-time (Fig. 7 and Fig. 10). Therefore, the impacts of increasing leading-time on the forecasted surface PM$_{2.5}$ concentrations in summer are mainly through its impacts on near-surface wind and precipitation and thus the dry and wet deposition rates.

Figure 10. Same as Fig. 5, but for precipitation.
In autumn, with the increasing leading-time, the total surface PM$_{2.5}$ mass increases in the 2-Day to 5-Day forecasts and then decreases in the 6-Day to 7-Day forecasts (Fig. 3). The overall change in total surface PM$_{2.5}$ mass with the leading-time is mainly due to the change in natural dust (Fig. 4). Part of the increase in surface PM$_{2.5}$ mass in the 2-Day to 5-Day forecasts is also from anthropogenic secondary particles, which is also partly related to condensation formation over the increase in existing particle surface area from natural dust. In the 2-Day to 5-Day forecasts, the change in anthropogenic primary particles is very small, although the PBL height and dry deposition rate change slightly with the leading-time. In the 5-Day to 7-Day forecasts, the change in the circulation pattern, particularly over the northern part, leads to a decrease in the surface concentrations of all the compositions. The contribution analysis of the transport process by Du et al. (2020) shows that the contributions of the transport process to the surface concentrations of all the compositions decrease from 5-Day to 7-Day (Fig. 11). Therefore, the impacts of increasing leading-time on the forecasted surface PM$_{2.5}$ concentrations in autumn are mainly through its impacts on the large-scale circulations, which can play a role in, on the one hand, transporting natural dust into the region and, on the other hand, ventilating the particles out of the region.
Figure 11. Monthly mean contributions of transport processes to surface concentrations of PM$_{2.5}$ compositions over the BTH region in October 2018 from the 2-Day to 7-Day forecasts.

4. Discussion

As analyzed above, this study focuses on investigating the leading-time impacts at a seasonal scale. However, the important role of leading-time in forecasting typical synoptic weather and haze events have also been reported by many studies (Gvozdikova
and Muller, 2021; Hochman et al., 2023; Keil et al., 2020; Ulpiani et al., 2022; Zhang et al., 2007). Here, a precipitation event (October 15th to 17th) and haze event (April 8th to April 10th) are selected to show the effect of forecast leading-time on typical events. As shown in Figure S4, the forecasted precipitation of the 2-Day forecast increases from 1500 UTC on October 15th, peaks at 0.45 mm h\(^{-1}\) at 0200 UTC on October 16th, and last until 1800 UTC on October 16th. As the leading-time increases, the intensity of precipitation decreases noticeably, and the time of peak precipitation shifts gradually. Ultimately, this precipitation event is entirely absent in the 7-Day forecast. The spatial distribution of precipitation at 850 hPa at 0200 UTC on October 16th, the time of peak precipitation, undergoes significant changes (Figure S5). These changes in intensity and spatial distribution of the predicted precipitation are caused by the movement and strength of the convergence of the wind field at 850 hPa with the increasing leading-time. The changes in precipitation weaken the contributions of wet depositions with increasing leading-time (Figure S4b). This ultimately leads to a considerable increase in PM\(_{2.5}\) concentrations, which rises almost threefold from the 2-Day (around 40 ug m\(^{-3}\)) to the 7-Day (roughly 120 ug m\(^{-3}\), Figure S4c).

As shown in Fig. S6, during the pollution event from April 8th to April 10th, the regional mean PM\(_{2.5}\) concentrations peak at 200 ug m\(^{-3}\) at 0000 UTC on April 9th in the 2-Day forecast. The PM\(_{2.5}\) concentrations increase gradually with the increase of leading-time, reaching over 400 ug m\(^{-3}\) in the 7-Day forecasts. It is obvious that natural dust is the dominant composition, accounting for more than 90% of the total PM\(_{2.5}\) mass. As the natural dust of the BTH region is mainly transported from northwestern China, this event is mainly caused by the long-range transported dust. In addition, the northwesterly winds
become stronger with the increase of leading-time (Figure S7), leading to enhanced long-range dust transport, which in turn contributes to the increasing trend in surface PM$_{2.5}$ concentrations in the BTH area.

Although the impacts of leading-time on a single event can also be analyzed as above, the numerical experiments designed in this study may not be appropriate for such analysis, because the analysis of a single event requires ensemble forecasts for each leading-time to eliminate the influence of initial condition and chaotic effect. Since the ensemble experiments for each leading-time for the entire four months are too computational expensive, they are not conducted in this study. Instead, this study focuses on the leading-time effect at a seasonal scale because the forecast experiment for each day can be taken as one ensemble member at seasonal scale so that the impacts of leading-time can be well assessed.

5. Conclusion

In this study, a series of forecasts with the leading-times from 2-Day to 7-Day are conducted for January, April, July, and October of 2018 over the BTH region using the WRF-Chem model. The seasonal characteristics of forecasting uncertainties in surface PM$_{2.5}$ concentrations associated with leading-time and its driving factors are investigated with these forecasts. On the regional average of the plain area of BTH, the forecasted highest surface PM$_{2.5}$ concentrations throughout the year occur in winter, with concentrations up to 55 ug m$^{-3}$, followed by those of approximately 35 ug m$^{-3}$ in spring and autumn. The forecasted surface PM$_{2.5}$ concentrations in summer reach the minimum of the year with a value of approximately 25 ug m$^{-3}$. 
With the increase of leading-time, the forecasted PM$_{2.5}$ concentrations change significantly and have obvious seasonal differences. Please note that these forecasting uncertainties in surface PM$_{2.5}$ concentrations are solely sourced from the forecasting uncertainties in meteorological fields due to the increasing leading-time. In general, the forecasting uncertainties of monthly mean surface PM$_{2.5}$ concentrations in the BTH region are the smallest (20%) in winter among the four seasons. In spring, the forecasted PM$_{2.5}$ concentrations increase from 32 $\mu$g m$^{-3}$ to 58 $\mu$g m$^{-3}$ with increasing leading-time, reflecting an uncertainty of ~80%. In summer, the forecasted surface PM$_{2.5}$ concentrations generally increase with the leading-time from 25 $\mu$g m$^{-3}$ in the 2-Day forecast to 35 $\mu$g m$^{-3}$, reflecting an uncertainty of ~40%. In autumn, the trend of forecasted surface PM$_{2.5}$ concentrations is similar to that in summer, and the uncertainty can reach 50%. Overall, the forecasting uncertainties in surface PM$_{2.5}$ concentrations can reach more than 80% solely due to the forecasting meteorological uncertainties with leading-times of 7-Day or even shorter.

The forecasting uncertainties in PM$_{2.5}$ composition and associated meteorological fields due to the increase of leading-time are further analyzed to explore the factors driving the seasonal characteristics of forecasting uncertainties in total surface PM$_{2.5}$ concentrations. In winter, anthropogenic primary particles dominate the total surface PM$_{2.5}$ mass and thus its forecasting uncertainties with leading-time. Therefore, the impact of leading-time in winter mainly results from its influence on PBL heights and hence the PBL mixing of anthropogenic primary particles. The uncertainties in near-surface wind and hence dry deposition rate also play a small role. In spring, the forecasting uncertainties in the total surface PM$_{2.5}$ mass mainly result from the impacts of leading-time on the large-scale northwesterly and thus the long-range transported natural dust. The increase in
transported natural dust can further provide more existing particle surface area for condensation production of anthropogenic secondary particles. In summer, the total mass of surface PM$_{2.5}$ is mainly occupied by anthropogenic primary and secondary particles. With increasing leading-time, the slight increase in the total surface PM$_{2.5}$ mass mainly results from the decrease in the dry and wet deposition rates, which is due to the reduction in the near-surface wind speed and precipitation rate. In autumn, the total surface PM$_{2.5}$ mass increases in the 2-Day to 5-Day forecasts and then decreases in the 6-Day to 7-Day forecasts. This is mainly due to the change in large-scale circulations with increasing leading-time, which eventually affects the long-range transport of natural dust and the ventilation of anthropogenic particles.

This study highlights the impacts of uncertainties in forecasted meteorological fields due to leading time on the forecasting uncertainties in surface PM$_{2.5}$ concentrations. The impacts have evident seasonal variation. Since the forecasting uncertainties in surface PM$_{2.5}$ concentrations can be significantly large due to leading-time, it is necessary to improve the forecast accuracy of key meteorological fields with an increase of leading-time before the accuracy of air quality forecasts can be truly made with a leading-time of a few days or even longer. According to previous studies, the intrinsic growth of errors in the underlying dynamical system and instabilities will contribute to the growth of forecasting uncertainties in meteorological fields by affecting some important processes, e.g., tropical cyclones, subtropical jet streams, blocking cases and convections (Dalcher and Kalnay, 1987; Emanuel and Zhang, 2016; Magnusson, 2017; Zhang et al., 2019). This may imply that the forecast accuracy of all these processes can affect the accuracy of forecasting surface PM$_{2.5}$ concentrations. In addition, in this study, the analysis of the
impact of leading-time on PM$_{2.5}$ concentrations forecast mainly focuses on the seasonal scale, and the impact of meteorological uncertainties are isolated while the impact of initial conditions and chaotic error are eliminated (more details are shown in Section 2.3). The forecasting uncertainties of surface PM$_{2.5}$ concentrations in typical synoptic weather and haze events due to the increase of leading-time can be even larger, which deserves further investigation.

**Data availability.** The release version of WRF-Chem can be downloaded from http://www2.mmm.ucar.edu/wrf/users/download/get_source.html. The updated USTC version of WRF-Chem can be downloaded from http://aemol.ustc.edu.cn/product/list/ or contact chunzhao@ustc.edu.cn. Additionally, code modifications will be incorporated into the release version of WRF-Chem in the future.

**Author contributions.** Qiuyan Du and Chun Zhao designed the experiments and conducted and analyzed the simulations. All authors contributed to the discussion and final version of the paper.

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Supporting materials for “Seasonal characteristics of forecasting uncertainties in surface PM2.5 concentration associated with leading-time over the Beijing-Tianjin-Hebei region”

Figure S1. Focused area of the analysis in this study. The shaded area denotes the area with a topography height of less than 500 m in the BTH region.
Figure S2. Spatial distribution of geopotential height and wind at 850 hPa from the ERA5 reanalysis dataset and the 2-Day forecast in January, April, July, and October of 2018.
Figure S3. Spatial distribution of temperature at 2m from the ERA5 reanalysis dataset and the 2-Day forecast in January, April, July, and October of 2018.
Figure S4. Time series of hourly precipitation (a), contributions of wet deposition on surface PM$_{2.5}$ concentrations (b) and the surface PM$_{2.5}$ concentrations (c) over the BTH region from the 2-Day to 7-Day forecasts during 0000 UTC October 15 to 0000 UTC October 17 of 2018.
Figure S5. Spatial distribution of hourly precipitation and wind fields at 850 hPa from the 2-Day to 7-Day forecasts at 0200 UTC on October 16 of 2018.
Figure S6. Time series of surface concentrations of PM$_{2.5}$ and PM$_{2.5}$ compositions over the BTH region from the 2-Day to 7-Day forecasts during 0000 UTC April 8 to 0000 UTC April 10 of 2018.
Figure S7. Spatial distribution of wind fields at 850 hPa from the 2-Day to 7-Day forecasts during 0000 UTC April 8 to 0000 UTC April 10 of 2018.
Figure S8. Spatial distribution of anthropogenic emissions of OC (a) and BC (b) for January, April, July, and October of 2018.