Aerosol-radiation feedback deteriorates the wintertime haze in North China Plain

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Abstract. Atmospheric aerosols scatter or absorb a fraction of the incoming solar radiation to cool or warm the atmosphere, decreasing surface temperature and altering atmospheric stability to further affect the dispersion of air pollutants in the planetary boundary layer (PBL). In the present study, simulations during a persistent and heavy haze pollution episode from 05 December 2015 to 04 January 2016 in the North China Plain (NCP) were performed using the WRF-Chem model to comprehensively quantify contributions of aerosol shortwave radiative feedback (ARF) to near-surface (around 15 m above the ground surface) $PM_{2.5}$ mass concentrations. The WRF-Chem model generally performs well in simulating the temporal variations and spatial distributions of air pollutants concentrations compared to observations at ambient monitoring sites in the NCP, and the simulated diurnal variations of aerosol species are also consistent with the measurements in Beijing. Additionally, the model simulates well the aerosol radiative properties, the downward shortwave flux, and the PBL height against observations in the NCP. During the episode, ARF deteriorates the haze pollution, increasing the near-surface $PM_{2.5}$ concentrations in the NCP by 10.2 $\mu g\ m^{-3}$ or with a contribution of 7.8% on average. Sensitivity studies have revealed that high loadings of $PM_{2.5}$ attenuate the incoming solar radiation reaching the surface to cool the low-level atmosphere, suppressing development of PBL, decreasing the surface wind speed, further hindering the $PM_{2.5}$ dispersion and consequently exacerbating the haze pollution in the NCP. Furthermore, when the near-surface $PM_{2.5}$ mass concentration increases from around 50 to several hundred $\mu g\ m^{-3}$, ARF contributes to the near-surface $PM_{2.5}$ by more than 20% during daytime in the NCP, substantially aggravating the heavy haze formation. However, when the near-surface $PM_{2.5}$ concentration is less than around 50 $\mu g\ m^{-3}$, ARF generally reduces the

42 near-surface $PM_{2.5}$ concentration due to the consequent perturbation of atmospheric dynamic

43 fields.

48

1 Introduction

Atmospheric aerosols, produced both naturally and anthropogenically, influence the radiative energy budget of the Earth's atmospheric system in many ways. They scatter or absorb a fraction of the incoming solar radiation to cool or warm the atmosphere, decreasing surface temperature and altering atmospheric stability (e.g., Ackerman, 1977; Jacobson, 1998, 2002). Also, they serve as cloud condensation nuclei (CCN) and ice nuclei (IN), thus modifying cloud optical properties and lifetime (e.g., Zhang et al., 2007; Li et al., 2008; 2009). Among those impacts, the scattering and absorption of solar radiation by aerosols and the associated feedbacks (hereafter referred to as aerosol-radiation feedback or ARF) not only constitute one of the main uncertainties in climate prediction (IPCC, 2007), but also substantially affect the atmospheric chemistry by perturbing the temperature profile and moistures, winds, and planetary boundary layer (PBL) stability (Boucher et al., 2013). Particularly, as a short-lived pollutant with uneven distribution and physical and chemical heterogeneities in the atmosphere, ARF varies by more than a factor of ten with location or time of emissions (Penner et al., 2010).

During wildfire with high loading absorbing aerosols, ARF has been reported to heat the atmosphere and cool the surface, and thence enhance the PBL stability (e.g., Grell et al., 2011; Fu et al., 2012; Wong et al., 2012). In addition, numerous studies have been performed to evaluate impacts of ARF of dust on the regional meteorology and climate (e.g., Perez et al., 2006; D. Zhang et al., 2009; Santese et al., 2010). Anthropogenic aerosols, dominated by scattering components, such as organics and sulfate, primarily attenuate the incoming solar radiation down to the surface, cooling the temperature of the low-level atmosphere to suppress the development of PBL and hinder the aerosol dispersion in the vertical direction (e.g., Fast et al., 2006; Vogel et al., 2009; Zhang et al., 2010). In addition, the temperature profile perturbation caused by ARF also alters cloud formation and development, possibly

74 causing the precipitation delay or decrease (e.g., Zhao et al., 2005; Koch and Del Genio, 2010;

75 Ding et al., 2013).

76   Rapid industrialization and urbanization in China have significantly elevated the

77 concentrations of aerosols or fine particulate matters ($PM_{2.5}$), causing frequent occurrence of

78 haze pollution, particularly during wintertime in North China (e.g., Zhang et al., 2013; Pui et

79 al., 2014). Guo et al. (2014) have elucidated the haze formation mechanism in China,

80 highlighting the efficient aerosol nucleation and growth during haze episodes. Moreover,

81 high loading aerosols during heavy haze episodes induce efficient ARF, encumbering the

82 PBL development and further deteriorating the haze pollution. It is worth noting that ARF

83 increases precursors for the aerosol nucleation and growth in the PBL, such as sulfuric and

84 organic gases, causing efficient aerosol nucleation and growth (Zhang et al., 2004; Guo et al.,

85 2014). Based on field measurements, recent studies have proposed that the high level of

86 $PM_{2.5}$ increases the stability of PBL due to ARF and further decrease the PBL height (PBLH),

87 consequently enhancing $PM_{2.5}$ concentrations ($[PM_{2.5}]$) (Quan et al., 2013; Petaja et al., 2016;

88 Yang et al., 2016; Tie et al., 2017; Ding et al., 2017). Online-coupled meteorology and

89 chemistry models have also been used to verify the impact of ARF on the PBLH and

90 near-surface $[PM_{2.5}]$ during haze episodes in Europe, Eastern China and Northern China

91 (Forkel et al., 2012; Z. Wang et al., 2014; Wang et al., 2015; Zhang et al., 2015; Gao et al.,

92 2015). However, the ARF impact on near-surface $[PM_{2.5}]$ varies, depending on the evaluation

93 time and location (Table 1). For example, the two-way coupled WRF-CMAQ system has

94 been employed to evaluate the ARF contribution to the haze formation in January 2013 over

95 the North China Plain (NCP), showing that ARF reduces the PBLH by 100 m and enhances

96 near-surface $[PM_{2.5}]$ by up to 140 µg m^{-3} in Beijing (J. Wang et al., 2014). Therefore, it is

97 still imperative to comprehensively quantify the ARF contribution to near-surface $[PM_{2.5}]$

98 under various pollution levels to provide the underlying basis for supporting the design and

99   implementation of emission control strategies.

100   In this study, simulations are performed using the Weather Research and Forecasting

101   model with Chemistry (WRF-Chem) to interpret the relationship between the near-surface

102   [$PM_{2.5}$] and the PBLH and further quantify the ARF contribution to near-surface [$PM_{2.5}$]

103   under various pollution levels. The model and methodology are described in Section 2.

104   Analysis results and discussions are presented in Section 3, and summary and conclusions are

105   given in Section 4.

106   ## 2   Model and methodology

107   ### 2.1   WRF-Chem model and configurations

108   The WRF-Chem model (Grell et al., 2005) with modifications by Li et al. (2010, 2011a,

109   b, 2012) is applied to evaluate effects of ARF on the wintertime haze formation in the NCP.

110   The model includes a new flexible gas phase chemical module, which can be used with

111   different chemical mechanisms, such as CBIV, RADM2, and SAPRC. In the study, the

112   SAPRC99 chemical mechanism is used based on the available emission inventory. For the

113   aerosol simulations, the CMAQ/models3 aerosol module (AERO5) developed by US EPA

114   has been incorporated into the model (Binkowski and Roselle, 2003). The wet deposition is

115   based on the method in the CMAQ module and the dry deposition of chemical species

116   follows Wesely (1989). The photolysis rates are calculated using the FTUV (fast radiation

117   transfer model) with the aerosol and cloud effects on photolysis (Li et al., 2005, 2011a).

118   It is worth noting that the most recent extension of ISORROPIA, known as ISORROPIA

119   II, has incorporated a larger number aerosol species (Ca, Mn, K salts) and is designed to be a

120   superset of ISORROPIA (Fountoukis et al., 2009). However, the ISORROPIA Version II

121   uses the exact same routines as ISORROPIA to compute the equilibrium composition, which

122   produces identical results as ISORROPIA when crustal species are not considered. Therefore,

123   the inorganic aerosols in this study are predicted using ISORROPIA Version 1.7, calculating

the composition and phase state of an ammonium-sulfate-nitrate-water inorganic aerosol in thermodynamic equilibrium with gas phase precursors (Nenes, 1998). In addition, a parameterization of sulfate heterogeneous formation involving aerosol liquid water (ALW) has been developed and implemented into the model, which has successfully reproduced the observed rapid sulfate formation during haze days (Li et al., 2017a). The sulfate heterogeneous formation from SO_2 is parameterized as a first order irreversible uptake by ALW surfaces, with a reactive uptake coefficient of 0.5×10^{-4} assuming that there is enough alkalinity to maintain the high iron-catalyzed reaction rate.

The OA module is based on the VBS approach with aging and detailed information can be found in Li et al. (2011b). The POA components from traffic-related combustion and biomass burning are represented by nine surrogate species with saturation concentrations (C*) ranging from 10^{-2} to 10^{6} μg m^{-3} at room temperature (Shrivastava et al., 2008), and assumed to be semi-volatile and photochemically reactive (Robinson et al., 2007). The SOA formation from each anthropogenic or biogenic precursor is calculated using four semi-volatile VOCs with effective saturation concentrations of 1, 10, 100, and 1000 μg m^{-3} at 298 K. The SOA formation via the heterogeneous reaction of glyoxal and methylglyoxal is parameterized as a first-order irreversible uptake by aerosol particles with an uptake coefficient of 3.7×10^{-3} (Liggio et al., 2005; Zhao et al., 2006; Volkamer et al., 2007).

A persistent air pollution episode from 05 December 2015 to 04 January 2016 in the NCP is simulated using the WRF-Chem model. During the study episode, the average hourly [$PM_{2.5}$] in the NCP are approximately 127.9 μg m^{-3}, within the fourth grade of National Ambient Air Quality Standards with [$PM_{2.5}$] between 115 and 150 μg m^{-3} (moderately polluted, Feng et al., 2016). The persistent and widespread haze pollution episode with high [$PM_{2.5}$] in the NCP provides a suitable case for observation analyses and model simulations to investigate ARF effect on haze pollution. Figure 1a shows the model simulation domain,

149 and detailed model configurations can be found in Table 2.

2.2 Aerosol radiative module

151   In the present study, Goddard shortwave module developed by Chou and Suarez (1999,

152   2001) is employed to take into account the ARF effect on the haze formation. The aerosol

153   radiative module developed by Li et al. (2011b) has been incorporated into the WRF-Chem

154   model to calculate the aerosol optical depth (AOD or τ_a), single scattering albedo (SSA or

155   ω_a), and the asymmetry factor (g_a).

156   In the CMAQ aerosol module, aerosols are represented by a three-moment approach

157   with a lognormal size distribution:

$$n(lnD) = \frac{N}{\sqrt{2\pi}ln\sigma_g}exp[-\frac{1}{2}(\frac{lnD-lnD_g}{ln\sigma_g})^2] \qquad (1)$$

159   Where D is the particle diameter, N is the number distribution of all particles in the

160   distribution, D_g is the geometric mean diameter, and σ_g is the geometric standard deviation.

161   To calculate the aerosol optical properties, the aerosol spectrum is first divided into 48 bins

162   from 0.002 to 2.5 μm, with radius r_i. The aerosols are classified into four types: (1)

163   internally mixed sulfate, nitrate, ammonium, hydrophilic organics and black carbon, and

164   water; (2) hydrophobic organics; (3) hydrophobic black carbon; and (4) other unidentified

165   aerosols. These four kinds of aerosols are assumed to be mixed externally. For the internally

166   mixed aerosols, the complex refractive index at a certain wavelength (λ) is calculated based

167   on the volume-weighted average of the individual refractive index. Given the particle size

168   and complex refractive index, the extinction efficiency (Q_e), ω_a and g_a are calculated

169   using the Mie theory at a certain wavelength (λ). The look-up tables of Q_e, ω_a and g_a are

170   established according to particle sizes and refractive indices to avoid multiple Mie scattering

171   calculation. The aerosol optical parameters are interpolated linearly from the look-up tables

172   with the calculated refractive index and particle size in the module.

173   The aerosol optical depth (AOD or τ_a) at a certain wavelength (λ) in a given

174 atmospheric layer k is determined by the summation over all types of aerosols and all bins:

175
$$\tau_a(\lambda, k) = \sum_{i=1}^{48} \sum_{j=1}^{4} Q_e(\lambda, r_i, j, k) \pi r_i^2 n(r_i, j, k) \Delta Z_k \tag{2}$$

176 where $n(r_i, j, k)$ is the number concentration of j-th kind of aerosols in the i-th bin. ΔZ_k is

177 the depth of an atmospheric layer. The weighted-mean values of σ and g are then

178 calculated by (d'Almeida et al., 1991):

179
$$\omega_a(\lambda, k) = \frac{\sum_{i=1}^{48} \sum_{j=1}^{4} Q_e(\lambda, r_i, j, k) \pi r_i^2 n(r_i, j, k) \omega_a(r_i, j, k) \Delta Z_k}{\sum_{i=1}^{48} \sum_{j=1}^{4} Q_e(\lambda, r_i, j, k) \pi r_i^2 n(r_i, j, k) \Delta Z_k} \tag{3}$$

180
$$g_a(\lambda, k) = \frac{\sum_{i=1}^{48} \sum_{j=1}^{4} Q_e(\lambda, r_i, j, k) \pi r_i^2 n(r_i, j, k) \omega_a(r_i, j, k) g_a(\lambda, r_i, j, k) \Delta Z_k}{\sum_{i=1}^{48} \sum_{j=1}^{4} Q_e(\lambda, r_i, j, k) \pi r_i^2 n(r_i, j, k) \omega_a(r_i, j, k) \Delta Z_k} \tag{4}$$

181 When the wavelength-dependent τ_a, ω_a, and g_a are calculated, they can be used in the

182 Goddard shortwave module to evaluate the ARF. Detailed information can be found in Li et

183 al. (2011b).

184 **2.3 Data and statistical methods for comparisons**

185 The model performance is validated using the available measurements in the NCP,

186 including AOD, SSA, PBLH, downward shortwave flux (SWDOWN), aerosol species, and

187 air pollutants. The daily AOD is retrieved from Terra- and Aqua- Moderate Resolution

188 Imaging Spectroradiometer (MODIS) level 2 products, with a resolution of 0.1°×0.1°. The

189 hourly SSA is calculated using the measurement of the turbidity meter at the National Center

190 for Nanoscience and Technology (NCNST), Chinese Academy of Sciences (116.33°E,

191 39.99°N) in Beijing (Figure 1b). The daily PBLH at 12:00 Beijing time (BJT) is diagnosed

192 from the radiosonde observation at a meteorological site (116.47°E, 39.81°N) in Beijing. The

193 SWDOWN is measured by CM-11 pyranometers at four sites from Chinese Ecosystem

194 Research Network (CERN) in the NCP (Liu et al., 2016). The hourly measurements of O_3,

195 NO_2, SO_2, CO and $PM_{2.5}$ concentrations have been released by the China's Ministry of

196 Ecology and Environment (China MEP) since 2013. The hourly submicron sulfate, nitrate,

197 ammonium, and organic aerosols are measured by the Aerodyne Aerosol Chemical

198 Speciation Monitor (ACSM) at NCNST. The primary organic aerosol (POA) and SOA

199 concentrations are obtained from the ACSM measurement analyzed using the Positive Matrix

200 Factorization (PMF). In addition, we have also analyzed the relationship between

201 near-surface [$PM_{2.5}$] and the PBLH retrieved from the Lidar measurement at the Institute of

202 Remote Sensing and Digital Earth (IRSDE), Chinese Academy of Sciences (116.38°E,

203 40.00°N) in Beijing (Figure 1b).

204 In the present study, the mean bias (*MB*), root mean square error (*RMSE*) and the index

205 of agreement (*IOA*) are used to assess the performance of WRF-Chem model simulations

206 against measurements. The detailed description can be found in Supplementary Information

207 (SI).

208

209 ## 3 Results and discussions

210 ### 3.1 Model performance

211 We first define the base simulation in which ARF is considered (hereafter referred to as

212 f_{base}), and results from f_{base} are compared to observations in the NCP. Generally, the

213 model simulates well the horizontal distributions and temporal variations of $PM_{2.5}$, O_3, NO_2,

214 and SO_2 mass concentrations against measurements in the NCP. Additionally, the model also

215 reasonably well reproduces the temporal profiles of the aerosol species compared to

216 observations in Beijing. The detailed model validation of air pollutants in the NCP and the

217 aerosol species in Beijing can be found in SI.

218 ### 3.1.1 Aerosol radiative properties simulations in NCP

219 Aerosol radiative forcing mainly depends on AOD, SSA, and asymmetry parameter (*g*).

220 The model validations of AOD and SSA are provided in this study to further evaluate the

221 aerosol radiative effect on the air pollution. The daily AOD at 550 nm, retrieved from Terra-

222 and Aqua- MODIS level 2 products, is compared with the simulation. Figure 2a shows the

scatter plot of the daily retrieved and simulated AOD averaged in the NCP from 05

December 2015 to 04 January 2016. The simulated daily average AOD correlates well with

the observation, with a correlation coefficient of 0.86. Generally, the retrieved and simulated

AOD increases with deterioration of the haze pollution, but the model considerably

underestimates the AOD against the observation. Figure 2b presents the Taylor diagram

(Taylor, 2001) to show the variance, bias and correlation of the simulated and retrieved AOD

from 05 December 2015 to 04 January 2016. There exists a good relationship between the

simulated and retrieved daily AOD during the study episode, with correlation coefficients

generally ranging from 0.5 to 0.9, and standard deviation mostly varying from 0.25 to 1.0.

Figure 3 shows the pattern comparison of the retrieved and simulated AOD averaged during

the simulation period. The model reasonably reproduces the AOD distribution compared to

the observations in the NCP, but considerably underestimates the AOD. The simulated and

retrieved AOD averaged in the NCP during the simulation period is 0.43 and 0.59,

respectively. It is worth noting that the simulated AOD is not only dependent on the column

aerosol content and constituent, but is also significantly influenced by the relative humidity

(RH) controlling the aerosol hydroscopic growth. Additionally, the satellite retrieved AOD is

subject to contamination by existence of clouds, and considering the high occurrence

frequency of clouds during haze days, the retrieved AOD is generally higher than the

simulation (Engstrom and Ekman, 2010; Chand et al., 2012; Grandey et al., 2013).

Aerosols are the mixture of absorbing and scattering constituents in the atmosphere.

Their radiative effect of cooling or warming the atmosphere relies on many parameters, and

SSA is one of the most important (Satheesh et al., 2010). Figure 4 depicts the comparison of

the measured and simulated diurnal profiles of SSA at NCNST in Beijing during the episodes.

The model performs reasonably in simulating the daily variation of SSA in Beijing, with an

IOA of 0.69 and a *MB* of 0.0, but the overestimation or underestimation is rather large. SSA

248 is the ratio of scattering to extinction, which is highly sensitive to the relative distribution of

249 scattering and absorbing aerosol constituents in the atmosphere, and the RH determining the

250 hygroscopic growth of aerosols. Therefore, the uncertainties of the simulated SSA probably

251 originated from the model biases of aerosol constituents and the RH.

252 **3.1.2 Downward solar radiation simulations in North China Plain**

253 Figure 5 presents the daily profiles of simulated and observed SWDOWN at ground

254 surfaces in Beijing, Jiaozhouwan, Luancheng, and Yuancheng from 05 December 2015 to 04

255 January 2016. The WRF-Chem model simulates well the daily variation of SWDOWN,

256 especially in Jiaozhouwan, Luancheng, and Yucheng, with *IOAs* around 0.90. The model is

257 subject to overestimating the SWDOWN against measurements, with *MB*s ranging from 6.3

258 to 86.2 W m^{-2}. The SWDOWN reaching the ground surface is very sensitive to the cloud

259 cover and optical thickness. However, the WRF-Chem model still has difficulties in

260 accurately predicting the cloud cover and optical thickness, which might constitute one of the

261 most important reasons for model biases of the SWDOWN. In addition, the horizontal

262 resolution used in simulations cannot adequately resolve the cumulus clouds, also causing

263 uncertainties in the simulations of the SWDOWN.

264 **3.1.3 PBLH simulations in Beijing**

265 Figure 6 shows the temporal variations of the observed and simulated PBLH at a

266 meteorological site in Beijing from 05 December 2015 to 04 January 2016. The average

267 PBLH at 12:00 BJT during the episode at the meteorological site is 465.2 m, with the

268 minimum of 101.8 m and the maximum of 1017.9 m, showing decreased PBLH during the

269 haze episode. In general, the WRF-Chem model tracks reasonably the daily variation of the

270 PBLH in Beijing, with an *IOA* of 0.70. However, the model has difficulties in reproducing

271 the observed very low PBLH, e.g., less than 200 m. The PBLH varies substantially with time

272 due to many factors including large-scale dynamics, cloudiness, convective mixing, and the

273 diurnal cycle of solar radiation (Sivaraman et al., 2013). Therefore, the simulation

274 uncertainties of meteorological conditions constitute the main reason for the simulation bias

275 of PBLH. For example, the overestimation of SWDOWN at 12:00 BJT (Figure 5a) probably

276 caused the overestimation of PBLH in Beijing.

277 In general, the simulated variations of SWDOWN, PBLH, aerosol radiative properties,

278 air pollutants ($PM_{2.5}$, O_3, NO_2, SO_2, CO) and aerosol species are in good agreement with

279 observations, indicating that the simulations of meteorological conditions, chemical processes

280 and the emission inventory used in the WRF-Chem model are reasonable, providing a

281 reliable basis for the further investigation.

282 **3.2 Relationship between near-surface [$PM_{2.5}$] and PBLH**

283 Figure 7 presents the scatter plot of the Lidar retrieved PBLH at IRSDE and

284 near-surface [$PM_{2.5}$] at a monitoring site close to IRSDE during daytime (08:00 ~ 17:00 LT)

285 from 08 January to 20 February 2014. The wind speeds (WSPD) at a meteorological site

286 close to IRSDE are shown by the color of the filled circles in Figure 7. Additionally,

287 near-surface [$PM_{2.5}$] during daytime are also subdivided into 20 bins with the interval of 25

288 $\mu g \ m^{-3}$. The PBLH as the bin of near-surface [$PM_{2.5}$] is assembled, and an average of PBLH

289 in each bin is calculated (Nakajima et al., 2001; Kawamoto et al., 2006), which is represented

290 by the rectangle in Figure 7. Generally, on average, when the PBLH decreases from 1500 m

291 to around 400 m, the near-surface [$PM_{2.5}$] increase from 10 to more than 200 $\mu g \ m^{-3}$. When

292 near-surface [$PM_{2.5}$] exceed 200 $\mu g \ m^{-3}$, the PBLH remains 400~500 m. Previous studies

293 have also reported the nonlinear relationship between the PBLH and near-surface [$PM_{2.5}$],

294 and proposed that increasing [$PM_{2.5}$] reduce the PBLH or the ARF is attributed to the PBLH

295 decrease (e.g., Petaja et al., 2016; Tie et al., 2017; Liu et al., 2018).

296 The PBLH is primarily determined by the wind shear in the vertical direction and the

297 thermal condition of ground surfaces. The occurrence of low near-surface [$PM_{2.5}$] generally

298 corresponds to efficient dispersions of $PM_{2.5}$ in horizontal and/or vertical directions. The

299 strong horizontal winds in the lower atmosphere not only disperse $PM_{2.5}$ emitted or formed

300 efficiently, but also intensify the wind shear in the vertical direction, increasing the PBLH

301 and facilitating the rapid vertical exchange of $PM_{2.5}$ in the PBL. When near-surface $[PM_{2.5}]$

302 are less than 50 μg m^{-3}, the PBLH exceeding 1000 m is observed, which is chiefly

303 determined by strong horizontal winds and less influenced by the ground thermal condition

304 during wintertime, and the observed average WSPD is about 2.4 m s^{-1}. The occurrence of

305 high near-surface $[PM_{2.5}]$ indicates that the lower atmosphere is stable or stagnant, with weak

306 horizontal winds and inactive convections, hindering the dispersion of $PM_{2.5}$ in the horizontal

307 and vertical directions. Additionally, as the horizontal winds become weak or calm, the wind

308 shear in the vertical direction is diminished and the PBLH is dominated by the ground

309 thermal condition. When near-surface $[PM_{2.5}]$ increase from 50 to around 200 μg m^{-3}, the

310 PBLH decreases from around 700 to 400 m, and the average WSPD decreases to 1.8 m s^{-1}.

311 However, the increased $PM_{2.5}$ reducing PBLH still cannot be fully attributed to ARF, which

312 is more likely caused by the decrease of winds or the formation of stagnant situations in the

313 low-level atmosphere. When near-surface $[PM_{2.5}]$ exceed 200 μg m^{-3}, the observed PBLH

314 fluctuates between 400 and 500 m with the average WSPD of around 1.0 m s^{-1}, and does not

315 exhibit continuous decrease with the increasing near-surface $[PM_{2.5}]$.

316 Under the stagnant situation with weak winds, the PBLH is more sensitive to the ground

317 thermal condition. Increasing aerosols or $PM_{2.5}$ in the low-level atmosphere attenuate the

318 SWDOWN to the ground surface and decrease the surface temperature (TSFC) and

319 turbulence kinetic energy, suppressing the PBL development and further enhancing

320 near-surface $[PM_{2.5}]$. Therefore, with near-surface $[PM_{2.5}]$ exceeding 200 μg m^{-3}, the inert

321 PBLH might be caused by the defect of the Lidar retrieved PBLH. The aerosol backscatter

322 signal received by Lidar is used to retrieve the PBLH. If the atmosphere is stable, the aerosols

323 near the maximal PBLH are subject to being confined in situ, and the retrieved PBLH is

324 generally the maximal one. Additionally, it is worth noting that the occurrence of the

325 wintertime severe haze pollution in the NCP is often accompanied with the high-level

326 convergence between 500 and 700 hPa, producing a persistent and strong sinking motion in

327 the mid-lower troposphere to reduce the PBLH and facilitate accumulation of air pollutants

328 (Wu et al. 2017; Ding et al., 2017). Therefore, a subsidence inversion appears in the lower

329 layer as a result of the air masses sinking in the middle-troposphere, restraining the PBL

330 development and determining the maximal PBLH. Hence, it is imperative to evaluate the

331 contribution of ARF to the PBLH and near-surface [$PM_{2.5}$].

332 ### 3.3 Sensitivity studies

333 The conceptual model about the ARF contribution to the heavy haze formation has been

334 established in previous studies (e.g., Tie et al., 2017; Liu et al., 2018). During wintertime,

335 under stagnant meteorological situations with weak winds and humid air, air pollutants are

336 subject to accumulation in the PBL, facilitating the formation of $PM_{2.5}$. Increasing $PM_{2.5}$ in

337 the PBL absorbs or scatters the incoming solar radiation to decrease the TSFC and facilitate

338 anomalous temperature inversion, subsequently suppressing the vertical turbulent diffusion

339 and decreasing the PBLH to further trap more air pollutants and water vapor to increase the

340 RH in the PBL. Increasing RH enhances aerosol hygroscopic growth and multiphase

341 reactions and augments the particle size and mass, causing further dimming and decrease of

342 the TSFC and PBLH. The whole process constitutes a positive feedback induced by the

343 aerosol radiation effect to enhance near-surface [$PM_{2.5}$], which has been proposed in many

344 studies (Quan et al., 2013; Petaja et al., 2016; Yang et al., 2016; Tie et al., 2017; Ding et al.,

345 2017; Liu et al., 2018). The noted positive meteorological condition feedback has also been

346 considered as the main reason for the near-surface $PM_{2.5}$ explosive growth (Zhong et al., 2018;

347 X. Y. Zhang et al., 2018).

348   To comprehensively evaluate the influence of ARF on near-surface [PM$_{2.5}$] during the

349   haze episode, a sensitivity study has been conducted, in which ARF is turned off (hereafter

350   referred as f_{rad0}). Therefore, the contribution of ARF to near-surface [PM$_{2.5}$] can be

351   determined by the difference between f_{base} and f_{rad0} (f_{base}- f_{rad0}). The most polluted

352   area in the NCP is first selected to verify the conceptual model of the ARF contribution to the

353   heavy haze formation, with the average near-surface [PM$_{2.5}$] during the haze episode

354   exceeding 150 μg m^{-3}. Figure 8 provides the temporal variation of near-surface [PM$_{2.5}$],

355   SWDOWN, TSFC, PBLH, and RH averaged in the selected area during the episode in f_{base}

356   and f_{rad0}. Apparently, ARF considerably decreases the solar radiation reaching the ground

357   surface and correspondingly lowers the TSFC (Figures 8b and 8c). Subsequently, the PBLH

358   is decreased and the surface RH is increased due to decreasing TSFC during daytime (Figures

359   8d and 8e). However, the variation trend of near-surface [PM$_{2.5}$], PBLH, TSFC and RH due

360   to ARF is not similar to that proposed in the conceptual model. During the haze development

361   stage, whether ARF is considered or not, the TSFC and RH exhibit an increasing trend,

362   showing the air mass originated from the south, and the PBLH does not consistently decrease

363   with increasing near-surface [PM$_{2.5}$]. Additionally, the ARF contribution to near-surface

364   [PM$_{2.5}$] is generally marginal during the haze development stage. During the haze maturation

365   stage, ARF commences to elevate near-surface [PM$_{2.5}$] appreciably. It is worth noting that,

366   even if ARF is not considered in f_{rad0}, the heavy haze pollution still occurs during the

367   episode. For example, from 17 to 20 December 2015, without ARF, near-surface [PM$_{2.5}$] still

368   continue to increase from around 30 to 300 μg m^{-3}, and fluctuate between 150 to 300 μg m^{-3}

369   until the occurrence of favorable meteorological conditions on 25 December. Hence,

370   according to the variation trend of near-surface [PM$_{2.5}$] with and without the ARF

371   contribution, the continuous accumulation of PM$_{2.5}$ during the haze episode is not primarily

372   caused by ARF, but predominantly induced by the stagnant meteorological conditions as well

373   as the massive air pollutants emissions in the NCP. Figure 9 presents the temporal variation

374   of AOD at 550nm averaged in the selected area during the episode in f_{base} and f_{rad0} to

375   evaluate the impact of ARF on AOD. Apparently, except from 8 to 11 December, the ARF

376   contribution to AOD is generally marginal, indicating that ARF does not play an important

377   role in the column-integrated aerosol abundance. Additionally, the considerable AOD

378   enhancement from 8 to 11 December is more likely caused by the substantial increase in RH

379   due to ARF, which facilitates aerosol hygroscopic growth to augment particle size and further

380   increases AOD. It is worth noting that the extinction of haze aerosols in the PBL also

381   decreases the photolysis to suppress the photochemistry, further hindering the secondary

382   aerosol formation to offset effects of ARF on near-surface [$PM_{2.5}$].

383   In order to quantitatively evaluate effects of ARF on near-surface [$PM_{2.5}$], which cannot

384   be reflected by the temporal variation of near-surface [$PM_{2.5}$], TSFC, PBLH and RH, an

385   ensemble method is used in this study. The daytime near-surface [$PM_{2.5}$] in the NCP during

386   the episode in f_{base} are first subdivided into 30 bins with an interval of 20 μg m^{-3}. The

387   SWDOWN, TSFC, PBLH, the near-surface WSPD, RH, and [$PM_{2.5}$] in f_{base} and f_{rad0} in

388   the same grid cell are assembled as the bin [$PM_{2.5}$], respectively, and an average of these

389   variables in each bin is calculated. Figure 10 shows the decrease of SWDOWN (%), TSFC

390   (^{o}C), PBLH (%), WSPD (m s^{-2}), and the increase of RH (%, not percentage change) and

391   near-surface [$PM_{2.5}$] contribution (%) caused by ARF as a function of bin [$PM_{2.5}$]. The

392   SWDOWN reaching the ground surface almost decreases linearly with the enhancement of

393   near-surface [$PM_{2.5}$]. When ARF is considered, aerosols in the atmosphere absorb or scatter

394   the incoming solar radiation, directly attenuating the radiation reaching the ground surface.

395   When near-surface [$PM_{2.5}$] exceed 200 μg m^{-3}, the SWDOWN at ground surfaces decreases

396   by more than 20% (Figure 10a). Moreover, the decrease of the SWDOWN correspondingly

397   lowers the TSFC and the decrease of the TSFC is generally proportional to near-surface

398 [$PM_{2.5}$], about $0.35\,^{\circ}C$ per 100 $\mu g\ m^{-3}$ $PM_{2.5}$ (Figure 10b). Interestingly, ARF also decreases

399 near-surface WSPD by about 0.1~0.2 $m\ s^{-1}$ with near-surface [$PM_{2.5}$] exceeding 80 $\mu g\ m^{-3}$

400 (Figure 10c). When severe air pollution occurs in the NCP during wintertime, atmospheric

401 convergence occurs in the PBL (Liao et al., 2015; Ding et al., 2017). However, the ARF

402 induced cooling in the low-level air generates a divergence in NCP, causing the decrease of

403 near-surface WSPD.

404   The PBLH is primarily determined by the atmospheric dynamic and thermal condition of

405 ground surfaces. Therefore, the decrease of WSPD and TSFC due to ARF subsequently

406 suppresses the PBL development and diminishes the PBLH (Figure 10d). When near-surface

407 [$PM_{2.5}$] are less than 250 $\mu g\ m^{-3}$, the PBLH decreases rapidly with increasing [$PM_{2.5}$]. When

408 the near-surface [$PM_{2.5}$] are between 250 $\mu g\ m^{-3}$ and 350 $\mu g\ m^{-3}$, the decrease of PBLH is

409 around 28%. With near-surface [$PM_{2.5}$] more than 350 $\mu g\ m^{-3}$, the decrease of PBLH exceeds

410 30%. As for ARF effect on water vapor in the PBL, the conceptual model has proposed that

411 the decreased PBL induced by ARF weakens the vertical exchange of water vapor or the

412 dispersion of water vapor is constrained by the shallow PBL (Tie et al., 2017; Liu et al.,

413 2018). However, Figure 11a shows that ARF decreases the near-surface water vapor content

414 slightly, by more than 0.1 $g\ kg^{-1}$ with near-surface [$PM_{2.5}$] exceeding 100 $\mu g\ m^{-3}$. During the

415 haze episode in the NCP, the abundant moisture in the PBL is mainly transported from the

416 south. The divergence due to cooling caused by ARF weakens the prevailing southerly wind

417 and decreases the moisture transport from the south, reducing the water vapor content in the

418 NCP. Considering that the RH is sensitive to the temperature with a constant water vapor

419 content, the ARF induced cooling still increases the near-surface RH (Figure 10e). When

420 near-surface [$PM_{2.5}$] exceed 300 $\mu g\ m^{-3}$, the RH is increased by more than 5%, so the heavy

421 haze generally causes the air to be more humid.

422   More $PM_{2.5}$ emitted or formed are trapped by a shallow PBL caused by ARF, and

423 increased RH promotes the aerosol hygroscopic growth and further multiphase reactions,

424 progressively enhancing near-surface $[PM_{2.5}]$ (Figure 10f). When near-surface $[PM_{2.5}]$ are

425 more than 50 µg m^{-3}, the contribution of ARF to near-surface $[PM_{2.5}]$ consistently increases

426 with the haze deterioration. When the severe haze occurs, i.e., near-surface $[PM_{2.5}]$ exceed

427 250 µg m^{-3}, more than 12% or 30 µg m^{-3} $PM_{2.5}$ is contributed by ARF. The simulated ARF

428 effects on near-surface $[PM_{2.5}]$ are generally comparable to those reported by previous

429 studies. Z. Wang et al. (2014) have shown that ARF increases the monthly $PM_{2.5}$

430 concentration by 10%-30% in Beijing-Tianjin-Hebei in January 2013. Using the WRF-Chem

431 model, Gao et al. (2015) have indicated that ARF increases the $PM_{2.5}$ concentration by 10-50

432 µg m^{-3} (2%-30%) over Beijing, Tianjin, and south Hebei from 10 to 15 January 2013, a

433 period with the simulated maximum hourly surface $PM_{2.5}$ concentration of more than 600 µg

434 m^{-3}. X. Zhang et al. (2018) have also quantified the aerosol-meteorology interaction effect on

435 $PM_{2.5}$ concentrations in China in 2014 using the WRF-Chem model, showing that the

436 increase of $PM_{2.5}$ concentrations associated with ARF is up to 16% in China. Other previous

437 studies have also confirmed ARF effect during the heavy haze pollution episode (Wang et al.,

438 2015; Zhang et al., 2015; Gao et al., 2016). However, when near-surface $[PM_{2.5}]$ are less than

439 50 µg m^{-3}, the contribution of ARF to near-surface $[PM_{2.5}]$ is negative, although ARF

440 decreases PBLH and increases RH. One of the possible reasons for the negative contribution

441 of ARF is perturbations of wind fields caused by the ARF induced cooling. Figure 11b

442 presents the average vertical velocity (the net velocity by combining updrafts and downdrafts)

443 below about 400 m in f_{rad0} as a function of near-surface $[PM_{2.5}]$. Apparently, when ARF

444 is not considered, the area with near-surface $[PM_{2.5}]$ less than 100 µg m^{-3} is generally

445 controlled by downward airflow, and vice versa for the area with near-surface $[PM_{2.5}]$ more

446 than 100 µg m^{-3}. The ARF induced cooling generally cause a downward motion in the PBL

447 (Figure 11c), which suppresses the upward motion in the area with near-surface $[PM_{2.5}]$ more

than 100 µg m^{-3} to enhance near-surface [PM$_{2.5}$], but accelerates the downward motion in the area with near-surface [PM$_{2.5}$] less than 100 µg m^{-3} to strengthen the divergence intensity, further decreasing near-surface [PM$_{2.5}$]. Countered by the decrease of PBLH and increase of RH, the ARF contribution becomes positive with near-surface [PM$_{2.5}$] exceeding 50 µg m^{-3}.

Figure 12 presents spatial distributions of the average near-surface PM$_{2.5}$ contribution due to ARF during the episode. The average near-surface PM$_{2.5}$ contribution caused by ARF in the NCP is 10.2 µg m^{-3} or 7.8%, with the maximum exceeding 40 µg m^{-3} in the south of Hebei. On average, the ARF contribution to near-surface [PM$_{2.5}$] is the most significant in Tianjin, about 17.6 µg m^{-3} or 10.3%, followed by Hebei (11.6 µg m^{-3} or 9.3%), Shandong (11.5 µg m^{-3} or 7.3%), Henan (11.2 µg m^{-3} or 7.7%), Anhui (7.7 µg m^{-3} or 7.4%), Beijing (7.3 µg m^{-3} or 6.9%), and Jiangsu (7.0 µg m^{-3} or 6.2%). It is noteworthy that the ARF contribution during the episode in North China is generally positive, but in its surrounding area the contribution becomes negative. At a large scale, when the air pollution occurs during wintertime in North China, the vertical motion over the polluted area generally shows an ascending-descending-ascending distribution from the surface to the middle level of the troposphere, and wind directions present a structure of convergence-divergence-convergence accordingly (Liao et al., 2015; Wu et al., 2017; Ding et al., 2017). ARF cools the low-level atmosphere and induces a downward motion, which suppresses the upward motion in the convergence area in North China to increase near-surface [PM$_{2.5}$], but accelerates the downward motion in the divergence area to decrease [PM$_{2.5}$].

Furthermore, when ARF is considered, near-surface [PM$_{2.5}$] over the East and South China Sea are also increased, with an enhancement less than 5 µg m^{-3} (about 3% to more than 15%). Considering the low near-surface [PM$_{2.5}$] over sea, the [PM$_{2.5}$] enhancement might be caused by the PM$_{2.5}$ transport from the continent. Figure 13 shows the spatial distribution of the TSFC and wind field variation caused by ARF averaged during the episode. Apparently,

473 ARF causes a widespread cooling effect in East China, and the cooling is the most significant

474 in the NCP, with the maximum TSFC decrease exceeding 1.5°C. The cooling effect in the

475 NCP induces a weak northerly wind, decreasing the prevailing southerly wind during the

476 haze episode (Figure 13). Additionally, the cooling effect over the continent also intensifies

477 the temperature contrast between land and sea, producing a secondary circulation to transport

478 the $PM_{2.5}$ from the continent to the East and South China Sea.

479

480 **4   Conclusions**

481   In the study, a persistent haze pollution episode in the NCP from 05 December 2015 to

482 04 January 2016 is simulated using the WRF-Chem model to verify the ARF contribution to

483 the haze formation. Generally, the model reproduces well the spatial distributions and

484 temporal variations of $PM_{2.5}$, O_3, NO_2, SO_2, and CO mass concentrations against observations

485 in the NCP. The calculated temporal variations of aerosol species are also consistent with the

486 ACSM measurement in Beijing, particularly with regard to the simulation of sulfate, nitrate,

487 and ammonium. Moreover, the model simulates reasonably well the variation of SWDOWN,

488 PBLH, and aerosol radiative properties during the episode, compared to the measurement.

489   Previous studies have established that a positive feedback induced by ARF causes the

490 heavy haze formation by modulating the PBL and RH. However, model results demonstrate

491 that during the haze development stage in the NCP, ARF does not dominate accumulation of

492 near-surface $[PM_{2.5}]$, while ARF considerably enhances near-surface $[PM_{2.5}]$ during the haze

493 mature stage.

494   Ensemble analyses of model results show that, during daytime, ARF attenuates

495 SWDOWN reaching ground surfaces efficiently, and correspondingly the TSFC

496 progressively decreases with increasing near-surface $[PM_{2.5}]$ in the NCP. The ARF induced

497 cooling generates a divergence in the low-level atmosphere in the NCP, lowering the

near-surface WSPD and decreasing the water vapor transport from the south. The decreased WSPD and TSFC caused by ARF hinder the PBL development and the PBLH decreases rapidly with increasing near-surface [$PM_{2.5}$]. Although the water content in the NCP is decreased slightly, the RH is still increased due to the ARF induced cooling. A shallow PBL and more humid air caused by ARF accelerate the $PM_{2.5}$ accumulation and secondary pollutant formation, facilitating heavy haze formation. The contribution of ARF to near-surface [$PM_{2.5}$] increases from 12% to 20% when near-surface [$PM_{2.5}$] increase from 250 to 500 $\mu g\ m^{-3}$. However, ARF decreases the $PM_{2.5}$ level with near-surface [$PM_{2.5}$] less than 50 $\mu g\ m^{-3}$.

The average near-surface $PM_{2.5}$ contribution of ARF during the episode in the NCP is 10.2 $\mu g\ m^{-3}$ or 7.8%. ARF aggravates the heavy haze formation in North China, but in its surrounding area ARF slightly mitigates the haze pollution. Generally, there is a structure of convergence-divergence-convergence over the polluted area of North China from the surface to the middle level of the troposphere. A downward motion is induced due to the widespread cooling effect of the low-level atmosphere caused by ARF, impeding the upward motion in the convergence area in North China to increase near-surface [$PM_{2.5}$], but accelerating the downward motion in the divergence area to decrease [$PM_{2.5}$].

Although the model performs generally well in simulating air pollutants, aerosol species and radiative properties, SWDOWN, and PBLH, the uncertainties from meteorological fields and emission inventory still have potentials to influence ARF evaluation. Particularly, further studies need to be conducted to improve the AOD simulations. In this study, ARF only considers the aerosol effect on the solar radiation, and the influence of longwave radiation also needs to be included. It is worth noting that modification of photolysis by aerosol scattering or absorbing solar radiation ultimately alters the atmospheric oxidizing capacity to influence the secondary aerosol formation, which potentially offsets ARF effect on the haze

523 ==pollution. Hence, further studies need to be performed to evaluate the effect of aerosol==

524 ==photolysis interaction on the haze pollution.== In addition, aerosols play an important role in the

525 cloud process serving as cloud condensation nuclei (CCN) and ice nuclei (IN). Therefore,

526 aerosol-cloud interactions (aerosol indirect effect) modify temperature and moisture profiles

527 and further influence precipitation, leading to potential effects on the atmospheric chemistry

528 (Wang et al., 2011). Future studies should be performed to investigate the feedbacks of the

529 aerosol indirect effect on the air pollutants.

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533 *Author contribution.* Guohui Li, as the contact author, provided the ideas and financial

534 support, developed the model code, verified the conclusions, and revised the paper. Jiarui Wu

535 conducted a research, designed the experiments, carried the methodology out, performed the

536 simulation, processed the data, prepared the data visualization, and prepared the manuscript

537 with contributions from all authors. Naifang Bei provided the treatment of meteorological

538 data, analyzed the study data, validated the model performance, and reviewed the manuscript.

539 Bo Hu provided the observation data used in the study, synthesized the observation, and

540 reviewed the paper. Suixin Liu, Meng Zhou, Qiyuan Wang, Zirui Liu, and Yichen Wang

541 provided the data and the primary data process, and reviewed the manuscript. Xia Li, Lang

542 Liu, and Tian Feng analyzed the initial simulation data, visualized the model results and

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547

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841   Table 1 Impact of ARF on near-surface [PM$_{2.5}$] in China

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Reference	Time	Location	Impact on [PM$_{2.5}$]
Z. Wang et al. (2014)	January 2013	Beijing-Tianjin-Hebei	+10~30%
J. Wang et al. (2014)	January 2013	North China Plain	Up to +140 μg m^{-3}
Gao et al. (2015)	10-15 January 2013	Beijing, Tianjin, and south Hebei	+10-50 μg m^{-3} (2-30%)
Wang et al. (2015)	7-11 July 2008	Beijing, Tianjin, Hebei, East Shanxi, West Shandong, and North Henan	+14%
Zhang et al. (2015)	January 2013	Henan, Hubei, Guangxi, and Sichuan	Maximum +69.3 μg m^{-3}
Ding et al. (2016)	December 2013	Eastern China and the Sichuan Basin	Up to +100 μg m^{-3}
Gao et al. (2016)	January 2010	Shijiazhuang	More than +20 μg m^{-3}
X. Y. Zhang et al. (2018)	December 2016	Beijing	around +84% of [PM$_{2.5}$] during cumulative explosive growth
Liu et al. (2018)	15-21 December 2016	North China Plain	+56 μg m^{-3}
X. Zhang et al. (2018)	2014	China	over +16% for the daily maximum [PM$_{2.5}$]
Zhong et al. (2018)	January 2013, February 2014, December 2015, and December 2016 to 10 January 2017	Beijing	Over +70% of [PM$_{2.5}$] during cumulative explosive growth

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848   Table 2 WRF-Chem model configurations.
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Region	East Asia
Simulation period	05 December 2015 to 04 January 2016
Domain size	400 × 400
Domain center	35°N, 114°E
Horizontal resolution	12 km × 12 km
Vertical resolution	35 vertical levels with a stretched vertical grid with spacing ranging from 30 m near the surface, to 500 m at 2.5 km and 1 km above 14 km
Microphysics scheme	WSM 6-class graupel scheme (Hong and Lim, 2006)
Cumulus scheme	Grell-Devenyi ensemble scheme (Grell and Devenyi, 2002)
Boundary layer scheme	MYJ TKE scheme (Janjić, 2002)
Surface layer scheme	MYJ surface scheme (Janjić, 2002)
Land-surface scheme	Unified Noah land-surface model (Chen and Dudhia, 2001)
Longwave radiation scheme	Goddard longwave scheme (Chou and Suarez, 2001)
Shortwave radiation scheme	Goddard shortwave scheme (Chou and Suarez, 1999)
Meteorological boundary and initial conditions	NCEP 1°×1° reanalysis data
Chemical initial and boundary conditions	MOZART 6-hour output (Horowitz et al., 2003)
Anthropogenic emission inventory	Developed by Zhang et al. (2009) and Li et al. (2017), 2012 base year, and SAPRC-99 chemical mechanism
Biogenic emission inventory	Online MEGAN model developed by Guenther et al. (2006)

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                                    Figure Captions

856   Figure 1 (a) WRF-Chem simulation domain with topography and (b) Beijing-Tianjin-Hebei
857   area. In (a), the blue circles represent centers of cities with ambient monitoring sites
858   in, and the size of blue circles denotes the number of ambient monitoring sites of
859   cities. In (b), the blue and red filled circles denote the NCNST and IRSDE site,
860   respectively, the red filled rectangle denotes the meteorological site. The red numbers
861   denote the CERN sites with the solar radiation measurement. 1: Beijing urban; 2:
862   Jiaozhouwan; 3: Yucheng; 4: Luancheng.

863   Figure 2 (a) Scatter plot of the MODIS retrieved and simulated daily AOD, (b) Taylor
864   diagram (Taylor, 2001) to present the variance, bias and correlation of the retrieved
865   and simulated daily AOD averaged in the NCP from 05 December 2015 to 04 January
866   2016.

867   Figure 3 Spatial distribution of (a) retrieved and (b) simulated AOD averaged from 05
868   December 2015 to 04 January 2016 in the NCP.

869   Figure 4 Comparison of measured (black dots) and predicted (red line) diurnal profiles of
870   SSA in Beijing from 05 December 2015 to 04 January 2016.

871   Figure 5 Comparison of measured (black dots) and predicted (red line) diurnal profiles of the
872   SWDOWN reaching the ground surface in (a) Beijing, (b) Jiaozhouwan, (c)
873   Luancheng, and (d) Yucheng from 05 December 2015 to 04 January 2016.

874   Figure 6 Comparison of predicted diurnal profile (red line) of PBLH from 05 December 2015
875   to 04 January 2016 with observations at 12:00 BJT in Beijing.

876   Figure 7 Scatter plot of the PBLH and near-surface [PM$_{2.5}$] at IRSDE site from 12 January to
877   20 February 2014. The black rectangle shows the bin average of PBLH. The color of
878   the filled circles denotes the WSPD at the meteorological site close to IRSDE in
879   Figure 1b.

880   Figure 8 Temporal variations of the average (a) near-surface [PM$_{2.5}$], (b) SWDOWN at the
881   ground surface, (c) TSFC, (d) PBLH, and (e) RH in the most polluted area in the NCP
882   with [PM$_{2.5}$] of more than 150 μg m^{-3} in f_{base} (red solid line) and f_{rad0} (blue solid
883   line) from 05 December 2015 to 04 January 2016.

884   Figure 9 Temporal variations of the average AOD at 550nm in the most polluted area in the
885   NCP with [PM$_{2.5}$] of more than 150 μg m^{-3} in f_{base} (red solid line) and f_{rad0} (blue
886   solid line) from 05 December 2015 to 04 January 2016.

887   Figure 10 Average (a) percentage decrease of SWDOWN at the ground surface, (b) decrease
888   of TSFC, (c) decrease of WSPD, (d) percentage decrease of PBLH, (e) increase of RH,
889   and (f) percentage contribution of near-surface [PM$_{2.5}$] caused by ARF, as a function
890   of the near-surface [PM$_{2.5}$] in the NCP during daytime from 05 December 2015 to 04
891   January 2016.

892   Figure 11 Average decrease of (a) near-surface water vapor content and (c) vertical velocity
893   below 400 m caused by ARF, and (b) average vertical velocity below 400 m in f_{rad0}
894   as a function of the near-surface [PM$_{2.5}$] in the NCP during daytime from 05
895   December 2015 to 04 January 2016.

896   Figure 12 Near-surface [PM$_{2.5}$] contribution caused by ARF, averaged from 05 December
897   2015 to 04 January 2016 in NCP.

898 Figure 13 TSFC and wind filed variations caused by ARF, averaged from 05 December 2015
899   to 04 January 2016 in NCP.

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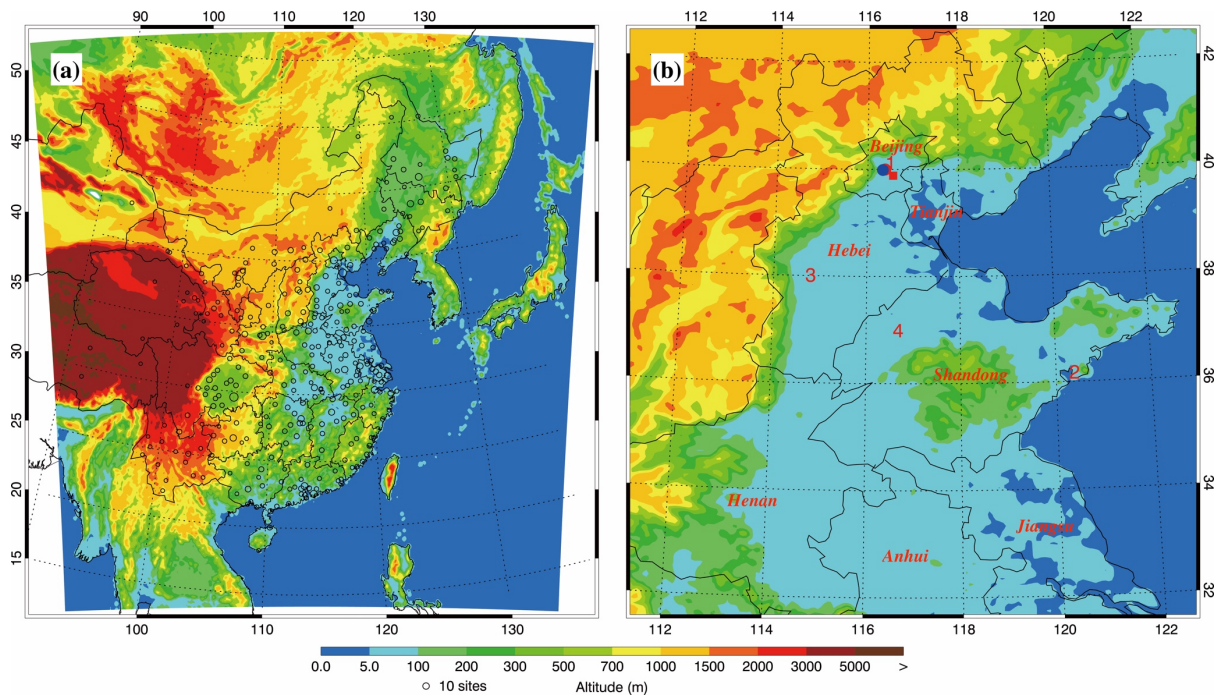
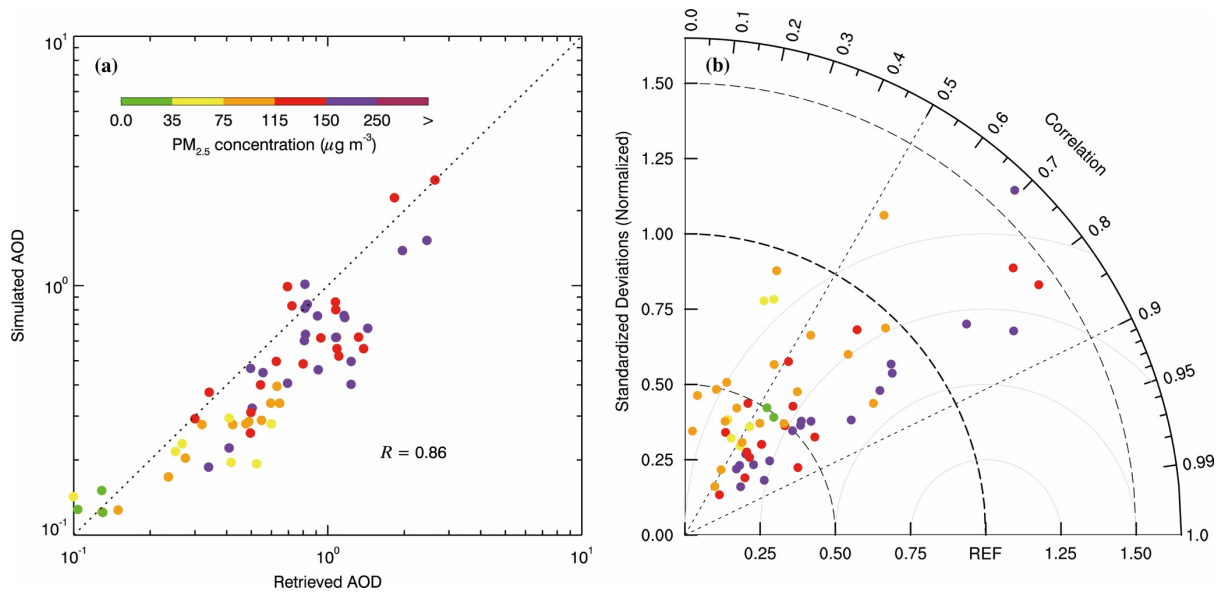


Figure 1 (a) WRF-Chem simulation domain with topography and (b) North China Plain. In (a), the blue circles represent centers of cities with ambient monitoring sites in, and the size of blue circles denotes the number of ambient monitoring sites of cities. In (b), the blue and red filled circles denote the NCNST and IRSDE site, respectively, and the red filled rectangle denotes the meteorological site. The red numbers denote the CERN sites with the solar radiation measurement. 1: Beijing urban; 2: Jiaozhouwan; 3: Yucheng; 4: Luancheng.

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920 Figure 2 (a) Scatter plot of the MODIS retrieved and simulated daily AOD, (b) Taylor
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922 simulated daily AOD averaged in the NCP from 05 December 2015 to 04 January 2016.
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Figure 3 Spatial distribution of (a) retrieved and (b) simulated AOD averaged from 05
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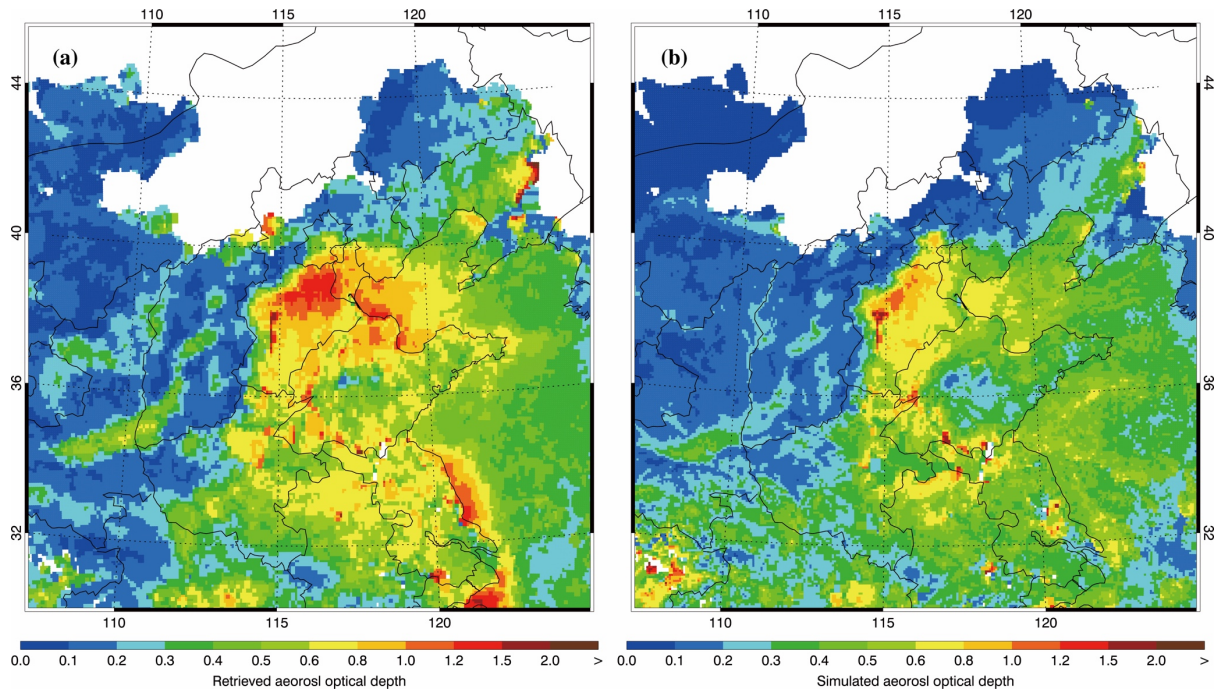
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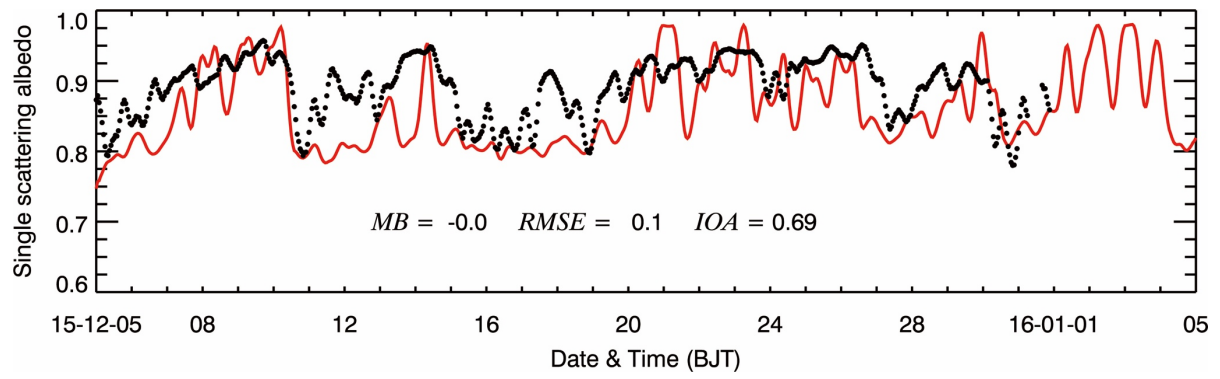
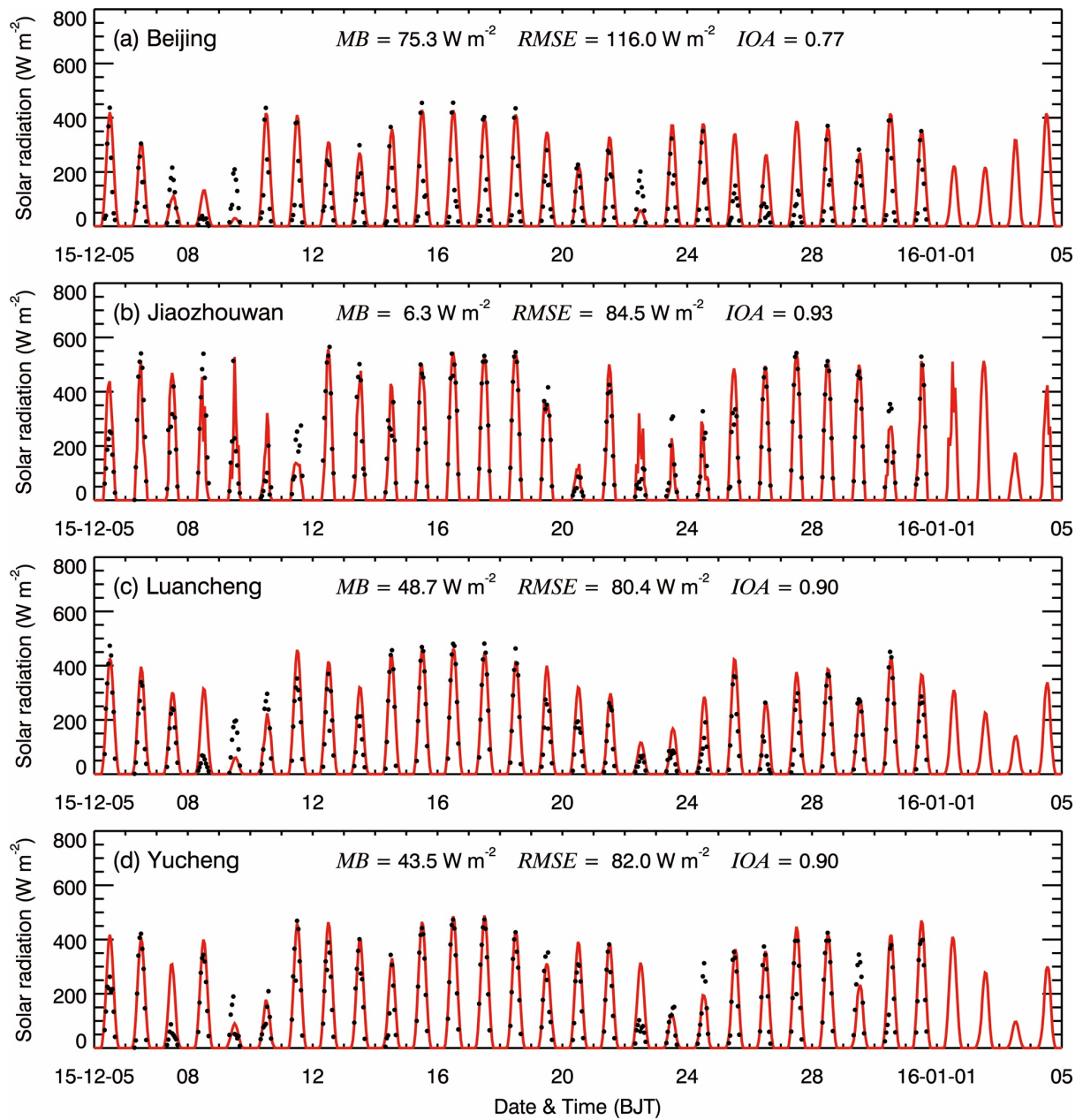


Figure 4 Comparison of measured (black dots) and predicted (red line) diurnal profiles of SSA in Beijing from 05 December 2015 to 04 January 2016.

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948 Figure 5 Comparison of measured (black dots) and predicted (red line) diurnal profiles of the
949 SWDOWN reaching the ground surface in (a) Beijing, (b) Jiaozhouwan, (c) Luancheng, and
950 (d) Yucheng from 05 December 2015 to 04 January 2016.
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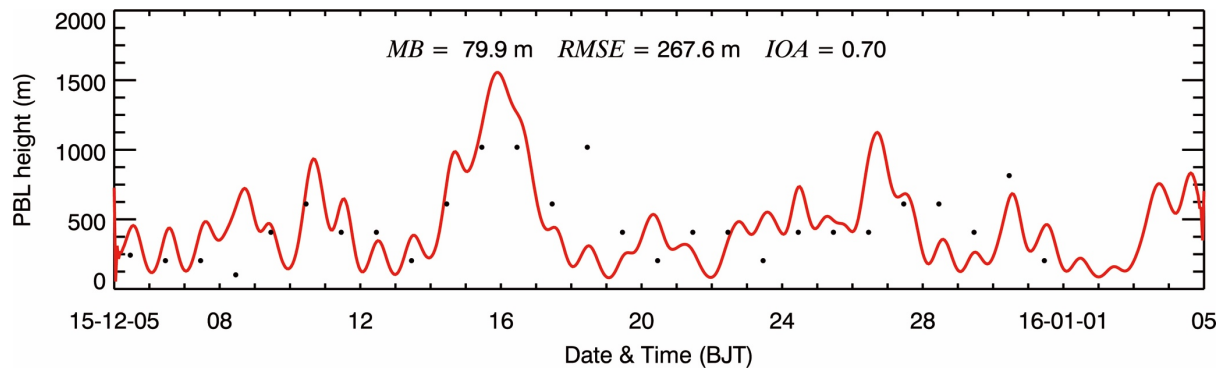
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958 Figure 6 Comparison of predicted diurnal profile (red line) of PBLH from 05 December 2015
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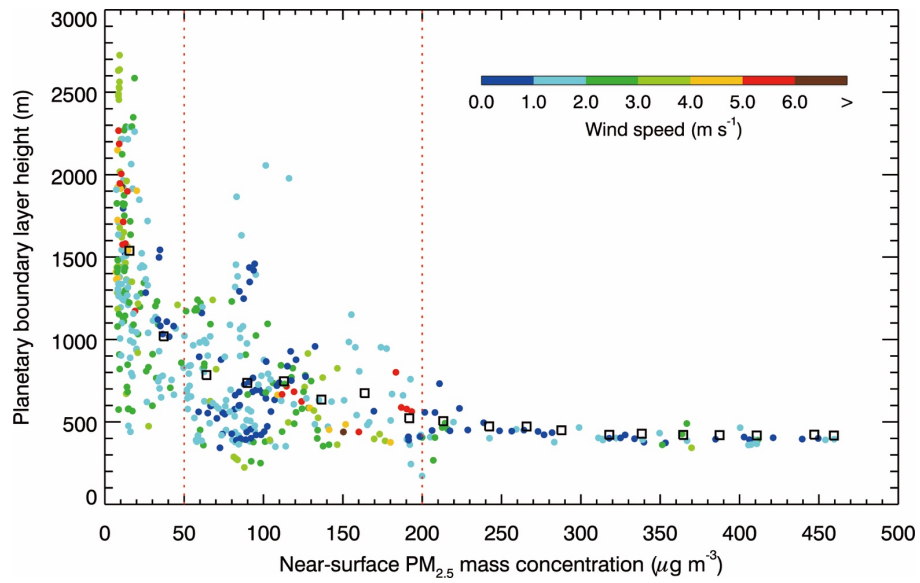
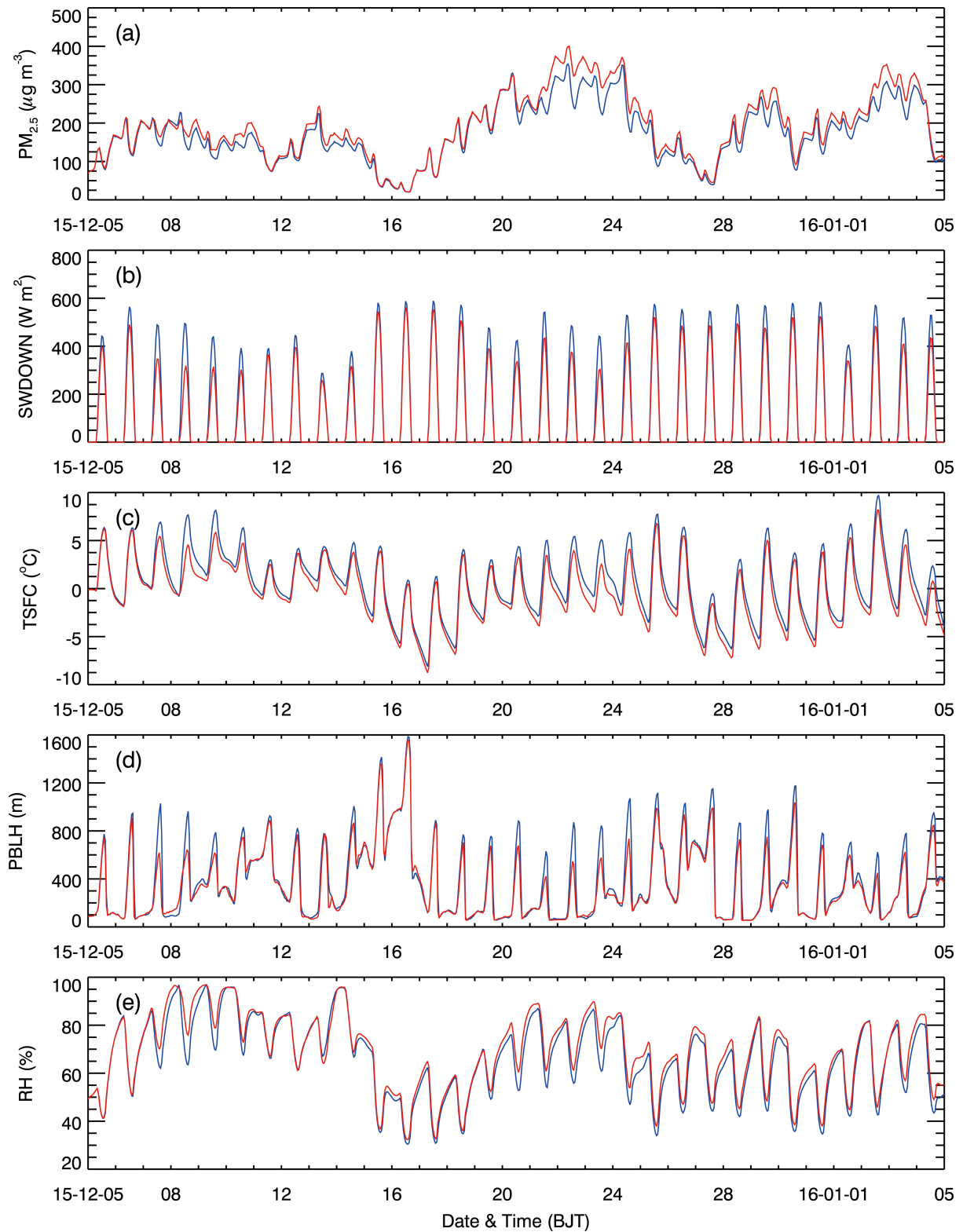

967 Figure 7 Scatter plot of the PBLH and near-surface [PM$_{2.5}$] at IRSDE site from 12 January to
968 20 February 2014. The black rectangle shows the bin average of PBLH. The color of the
969 filled circles denotes the WSPD at the meteorological site close to IRSDE in Figure 1b.

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977 Figure 8 Temporal variations of the average (a) near-surface [PM$_{2.5}$], (b) SWDOWN at the
978 ground surface, (c) TSFC, (d) PBLH, and (e) RH in the most polluted area in the NCP with
979 [PM$_{2.5}$] of more than 150 μg m^{-3} in f_{base} (red solid line) and f_{rad0} (blue solid line) from
980 05 December 2015 to 04 January 2016.

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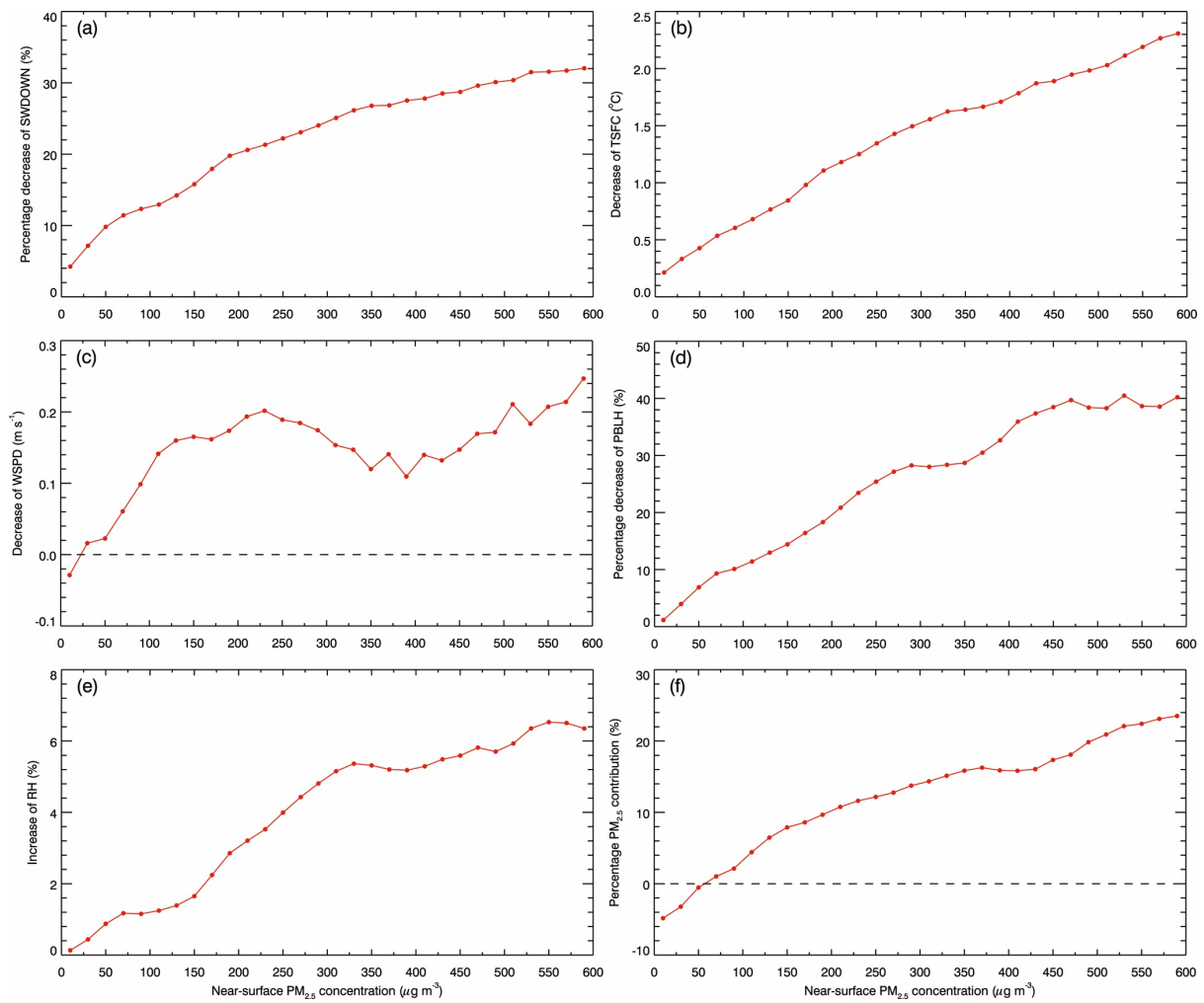
Figure 9 Temporal variations of the average AOD at 550nm in the most polluted area in the NCP with [PM$_{2.5}$] of more than 150 μg m^{-3} in f_{base} (red solid line) and f_{rad0} (blue solid line) from 05 December 2015 to 04 January 2016.

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995 Figure 10 Average (a) percentage decrease of SWDOWN at the ground surface, (b) decrease
996 of TSFC, (c) decrease of WSPD, (d) percentage decrease of PBLH, (e) increase of RH, and (f)
997 percentage contribution of near-surface [PM$_{2.5}$] caused by ARF, as a function of the
998 near-surface [PM$_{2.5}$] in the NCP during daytime from 05 December 2015 to 04 January 2016.
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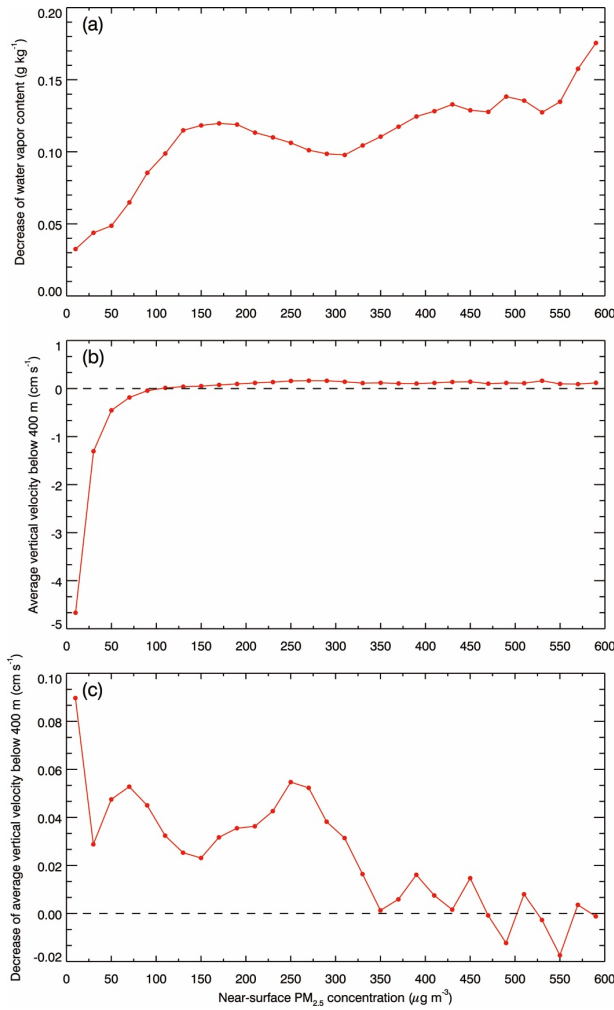
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1006 Figure 11 Average (a) decrease of water vapor content and (c) decrease of average vertical
1007 velocity below 400 m caused by ARF, and (b) average vertical velocity below 400 m in
1008 f_{rad0} as a function of the near-surface [PM$_{2.5}$] in the NCP during daytime from 05
1009 December 2015 to 04 January 2016.
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Figure 12 Near-surface [PM$_{2.5}$] contribution caused by ARF, averaged from 05 December 2015 to 04 January 2016 in the NCP.

Figure 13 TSFC and wind filed variations caused by ARF, averaged from 05 December 2015 to 04 January 2016 in the NCP.