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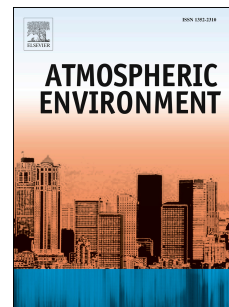
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Journal Pre-proof

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Journal Pre-proof

1 High Spatial Resolution WRF-Chem Model over Asia: Physics and 2 Chemistry Evaluation

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13 Abstract

14 The representation of air quality and meteorology over Asia remains challenging for chemical
15 transport models as a result of the complex interactions between the East Asian monsoons and
16 the large uncertainty (in space and time) of the high anthropogenic emissions levels over the
17 region. High spatial resolution models allow resolving small-scale features induced by the
18 complex topography of this region. In this study, the Weather Research and Forecasting
19 model with Chemistry (WRF-Chem) was used to simulate the spatial and seasonal variability
20 of main physical and chemical variables over Asia for the year 2015 at 8-km horizontal
21 resolution to enable resolving small-scale features induced by the region complex topography.
22 The simulated atmospheric composition was evaluated against satellite retrievals (MOPITT,
23 IASI+GOME2, MODIS and OMI) in addition to ground-based observations in China for the
24 year 2015, while the meteorological variables were evaluated by several observational-based
25 datasets (ERA5, CRU, MODIS, MTE). Results showed low to moderate seasonal biases for
26 major meteorological variables, i.e. air temperature, relative humidity, precipitation, latent
27 heat, sensible heat and snow cover fraction. Overall, WRF-Chem reproduced well the spatial
28 and seasonal variability of lowermost tropospheric ozone content, total column carbon
29 monoxide and aerosol optical depth, while large discrepancies were found for tropospheric
30 nitrogen dioxide content, mainly during the warm season. In consistency with previous
31 studies, the different biases between model-simulated and satellite-retrieved values can be
32 mainly attributed to i) the large uncertainties in anthropogenic and natural nitrogen oxides
33 emission estimates, as well as dust and sea salt emissions in the case of aerosol optical depth,
34 and ii) some coarse parameterizations used to reproduce main small-scale features (e.g.
35 meteorology, chemical processes, dry deposition to vegetation). Compared to ground-based
36 observations, the WRF-Chem model reproduced well the mean annual cycle of surface
37 nitrogen dioxide, ozone and fine particles concentrations in all seasons across China. Our
38 results suggest that WRF-Chem provides reliable spatio-temporal patterns for most of the
39 meteorological and chemical variables, adding thus confidence to its applicability in the
40 context of air pollution risk assessment to human and ecosystems health.

41 **Keywords:** Asia, satellite, regional climate model, remote sensing, WRF-Chem

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43 1. Introduction

44 China and India are the two most populous countries of the world (~ 2.8 billion people) and
45 have experienced a rapid growth in industrial, transportation, urbanization and agricultural
46 activities in recent years (Kumar et al., 2015; Zhang et al., 2016a). This explosive economic
47 growth has significantly increased anthropogenic emissions of several trace gases and
48 aerosols over Asia in the last decades (Kumar et al., 2015; Zhang et al., 2016b; Lefohn et al.,
49 2017), with China becoming the largest emitter of air pollutants worldwide (Liu et al., 2015;
50 Quéré et al., 2015; Boden et al 2017; Wang et al., 2017a).

51
52 The increasing air pollution levels pose health risks to billions of people (Zhong et al., 2016):
53 in fact, in major metropolitan agglomerations across Asia (e.g. Bangladesh, China, India and
54 Nepal) surface ozone (O₃) concentrations regularly exceed the ambient air quality standard of
55 100 ppb as hourly value (Wang et al., 2017a), while annual PM_{2.5} concentrations exceed 100
56 µg m⁻³ (Zhang and Cao, 2015; Venkataraman et al., 2018). These high levels of air pollutants
57 produce acute and chronic effects on population including premature mortality due to cancer,
58 respiratory and cardio-vascular diseases (Lelieveld et al., 2015; Liu et al., 2016; Cohen et al.
59 2017; Krishna et al., 2017; Burnett et al., 2018) and reduced life expectancy (Apte et al.,
60 2018). In addition, forests and crops are remarkably affected by high pollution levels: in
61 particular, effects on plants include, among others, yield (Tang et al., 2013; Tai et al., 2014;
62 Sicard et al., 2016a; Tian et al., 2016; Feng et al., 2019) and biomass decline (Wittig et al.,
63 2009; Feng et al., 2015; Li et al., 2017). Krishna et al. (2017) estimated that air pollution
64 contributes to 13-22% of all deaths in South Asia. Other studies showed that for the year 2015
65 air pollution led to 4.5 million premature deaths and more than half occurring in China and
66 India (Landrigan et al., 2017; Giani et al., 2020), with fine particulate matters (PM_{2.5}) causing
67 around 1 million premature deaths every year (Cohen et al., 2017; Li et al., 2018; Burnett et
68 al., 2018).

69
70 For these reasons, monitoring of air quality plays a pivotal role to preserve human and
71 ecosystem health; in this regard, in 2013, the State Council of China issued the Air Pollution
72 Prevention and Control Action Plan (Wang et al., 2018), while India introduced policies and
73 National Clean Air Programme (Sagar et al., 2016) to provide a framework for air quality
74 monitoring in order to mitigate the air pollution and attain air quality standards (Sagar et al.,
75 2016; Goldemberg et al., 2018; Wang et al., 2018). In addition to surface measurements,
76 chemical transport models (CTMs) represent a valuable tool to predict formation/removal of
77 air pollutants and their transport, and provide air quality information over remote regions or
78 scarcely populated areas where measurements are not available (Sicard et al., 2017).
79 However, before providing any assessment of impact of air pollution on human and
80 vegetation health, it is mandatory assessing how well models perform in reproducing the
81 spatio-temporal variability of both physical and chemical variables.

82
83 In general, regional CTMs have been found able to reproduce observed spatial pattern of air
84 pollutants and their seasonal changes (Spiridonov et al., 2019; Li et al., 2018; Liu et al., 2018)
85 in both North America and Europe (discussed below). However, regional chemistry models
86 still reproduce poorly the observed spatial pattern of main air pollutants over the Asian region
87 because of (i) complex landscape ranging from high elevations of Himalayan plateau to
88 megacities of Easter China; (ii) large uncertainties in removal of trace gases through dry
89 deposition to vegetation associated to a mosaic land cover (Monks et al., 2015), ranging from
90 tropical rainforest to boreal forest and semi-arid or desert area; (iii) widely-varying climate
91 system characterized by a summer monsoon system and (iv) large uncertainties in

92 anthropogenic emissions (e.g. Amnuaylojaroen et al., 2014; Jena et al., 2015). Consequently,
93 remote sensing data represent a unique opportunity to evaluate the spatio-temporal
94 distribution of air pollutants simulated by regional CTMs (Tuccella et al., 2012; Crippa et al.,
95 2016, 2017; Georgiou et al., 2018; Crippa et al., 2019). China adopted in 2012 the Ambient
96 Air Quality Standard for human health protection, and started reporting hourly observations of
97 main air pollutants from about 1,500 monitoring stations at countrywide (MEP, 2012). These
98 monitoring stations offers an unprecedented way to evaluate model-simulated surface
99 concentrations of air pollutants.

100
101 The coupled Weather Research and Forecasting model coupled with Chemistry (WRF-Chem)
102 model is widely used for regional air quality simulation worldwide and validated against
103 ground-based observations, e.g. over North America (McKeen et al., 2005; Chuang et al.,
104 2011; Archer-Nicholls et al., 2014; Yahya et al., 2015; Zhang et al., 2017), Europe
105 (Schürmann et al., 2009; Solazzo et al., 2012; Tuccella et al., 2012; Ritter et al., 2013;
106 Karlický et al., 2017; Werner et al., 2017; Spiridonov et al., 2019; Visser et al., 2019) and
107 South or East Asia (Tie et al., 2007; Kumar et al., 2012a,b; Gao et al., 2014; Zhong et al.,
108 2016; Zhang et al., 2016; Sharma et al., 2017; Li et al., 2018; Xu et al., 2018; Reddington et
109 al., 2019). A few studies reported the WRF-Chem performances over East Asia (Zhang et al.,
110 2016; Zhong et al., 2016) and South Asia (Kumar et al., 2012a,b; Sharma et al., 2017) for air
111 pollutants and meteorology, however the coarse resolution and lack of ground observations
112 limit the model skill to reproduce small-scale processes (e.g. Crippa et al., 2017; Chen et al.,
113 2019). In this regard, the new generation of high spatial resolution reanalysis offers a unique
114 opportunity to run CTMs at very high spatial resolutions over the South-East Asia.

115
116 The aim of this paper is to conduct a WRF-Chem simulation of meteorological fields and air
117 pollutants, and evaluate the model performance over a large area covering India and China.
118 The novelty is related to the availability of ground observations in China to validate the WRF-
119 Chem outputs. Therefore, we performed the simulations at fine resolution to allow the model
120 to reproduce well the local variability of climatic and chemical parameters. Despite ground-
121 based observations are amongst the most accurate and reliable datasets to evaluate regional
122 climate models, the lack of spatial representativeness of air quality monitoring stations
123 (Beelen et al., 2009; Sicard et al., 2016b) limits the model evaluation to the regions covered
124 by data. To overcome this gap in spatial heterogeneity, and assess the ability of the WRF-
125 Chem model to reproduce regional patterns of trace gases, we firstly compare simulated data
126 with satellite-based measurements; successively, we use recent ground observations across
127 China to validate surface concentrations of air pollutants.

128 129 **2. Materials and Methods**

130 **2.1. WRF-Chem model**

131 The WRF model is a limited-area, non-hydrostatic, terrain-following eta-coordinate
132 mesoscale model (Skamarock et al., 2008). This model has been further developed to include
133 various gas-phase chemistry and aerosol mechanisms creating the coupled chemistry-climate
134 WRF-Chem model (Grell et al., 2005). The WRF model system offers multiple options for
135 various physical packages (Skamarock et al., 2008). The dynamical core used in this work is
136 the Advanced Research Weather Research and Forecasting model (Tab. 1); we used a single-
137 moment 6-class scheme to resolve the microphysics (Hong et al., 2006) and the Rapid
138 Radiative Transfer Model for GCMs (RRTMG) for the shortwave and longwave radiation
139 (Iacono et al., 2008). Convective precipitation and cumulus parameterization were resolved

140 with the new Tiedtke scheme (Zhang et al., 2011), the planetary boundary layer computations
141 were performed using the nonlocal K-profile Yongsei University parameterization (Hong et
142 al., 2006), while the exchange of heat, water and momentum between soil-vegetation and
143 atmosphere was simulated by the Unified Noah Land Surface Model (Chen and Dudhia,
144 2001).

145
146 Similarly to physical parameterizations, many different gas phase chemistry and aerosol
147 options are available in WRF-Chem. Gas-phase chemical reactions are calculated using the
148 chemical mechanism MOZART (*Model for OZone And Related chemical Tracers*) (Emmons
149 et al., 2010) whereas for the aerosols, to reduce the computational cost, we used the GOCART
150 (*Global Ozone Chemistry Aerosol Radiation and Transport*) bulk aerosol approach (Chin et
151 al., 2000). This set-up includes 85 gas-phase species, 12 bulk aerosol compounds, 39
152 photolysis and 157 gas-phase kinetic reactions.

153
154 Anthropogenic emissions are based on the EDGAR-HTAP (*Emission Database for Global*
155 *Atmospheric Research for Hemispheric Transport of Air Pollution*) global emission inventory
156 which includes diurnal cycle of emissions of gaseous pollutants such as SO₂, NO_x, CO, non-
157 methane volatile organic compounds (NMVOCs) and NH₃ as well as black carbon and
158 particulate matter from the following source sectors: aviation, shipping, agriculture, power
159 generation, industrial non-power, land transport and residential energy use (Janssens-
160 Maenhout et al., 2015). This dataset is available at 0.1°×0.1° horizontal resolution for the year
161 2010, with no year adjustments. Fire emissions are provided using the FINN (Fire INventory
162 from NCAR) inventory (Wiedinmyer et al., 2011). This dataset provides estimates of trace
163 gases and particles emitted by open biomass burning at ~1 km resolution (Wiedinmyer et al.,
164 2011). Biogenic emissions are calculated online using the MEGAN (Model of Emissions of
165 Gases and Aerosols from Nature) model (Guenther et al. 2012), dust emissions are estimated
166 online using the GOCART model (Ginoux et al., 2001), whereas sea-salt emissions are
167 calculated using the method by Gong (2003). Anthropogenic dust emissions (e.g. re-
168 suspended road dust) are not included.

169
170 In addition, MOZART-4/Goddard Earth Observing System Model version 5 (GEOS-5) data
171 were used for chemical and aerosol boundary conditions. The MOZART-4 data is a model
172 outputs dataset available at a horizontal grid resolution of 1.9°×2.5° every 6 h and is driven by
173 the National Aeronautics and Space Administration (NASA). The initial and boundary
174 meteorological conditions (including time varying sea surface temperature), required to run
175 the model, are provided by the European Centre for Medium-range Weather Forecast
176 (ECMWF) re-analysis project ERA5, with a horizontal resolution of about 31 km every 3
177 hours.

178
179 In this study we used WRF-Chem (v3.9) to simulate meteorology and air quality from 20th
180 December 2014 to 31st December 2015 using the first 10 days as spin up. We conducted a
181 fully free running simulation (i.e. without nudging) for the entire year 2015. The model
182 domain is projected on a Lambert conformal grid (780 x 690 grid cells) with a horizontal grid
183 resolution of 8 km with 30 vertical levels extending from the surface up to 50 hPa. A
184 synthesis of parameterizations and input data used in this study is given in Tab. 1.

185

186 **2.2. Datasets for model evaluation**

187 To evaluate the spatio-temporal patterns of simulated surface air temperature, relative
188 humidity and precipitation, we compared model results against Climatic Research Unit (CRU)

189 observed data, a monthly high-resolution ($0.5^\circ \times 0.5^\circ$) gridded dataset over land areas (Mitchell
190 and Jones, 2005). In addition to CRU, which can be regarded as an independent dataset, for
191 the evaluation of physical variables, we also compared simulated data against the boundary
192 conditions used to drive the model (i.e. ERA5); this allows assessing if a bias has been
193 introduced into the model by its forcing (Mooney et al., 2013) or is mainly due to poor
194 representation of some physical processes within the model (Tang et al., 2017). In addition, in
195 order to remove any possible bias in surface temperature related to the difference in
196 topography between the coarse reference data and the finer model output, we downscaled
197 both CRU and ERA5 temperature to our domain using a dry-adiabatic lapse. Sensible and
198 latent heat fluxes were evaluated using the Machine Tree Ensemble (MTE) a $0.5^\circ \times 0.5^\circ$
199 gridded dataset (Jung et al., 2011) created by upscaling eddy covariance measurements
200 collected around the world (Jung et al. 2009, 2011). This dataset has been widely employed to
201 evaluate the performances of land surface models, including NOAH-MP, a land surface
202 scheme often used within WRF (Ma et al., 2017). We used the Moderate Resolution Imaging
203 Spectro-radiometer (MODIS) product from the NASA Terra Satellite to evaluate the snow
204 cover fraction (Hall et al., 2010); this dataset provides monthly data with a resolution of 0.05°
205 $\times 0.05^\circ$ (Hall and Riggs, 2015).

206 We have evaluated the WRF-Chem simulations of NO_2 concentrations against satellite
207 retrievals from the Ozone Monitoring Instrument (OMI), flying aboard NASA's EOS-Aura
208 satellite. This instrument measures the radiation backscattered by the Earth's atmosphere and
209 surface, and provides the daily global retrievals of several trace species and aerosols with a
210 spatial resolution of $13 \text{ km} \times 24 \text{ km}$ at nadir (Boersma et al., 2011). To evaluate the ability of
211 the model to reproduce reliable spatial and seasonal NO_2 estimates, we compared the
212 tropospheric NO_2 content simulated by WRF-Chem with spatial resolution of OMI data
213 available from KNMI (Royal Netherlands Meteorological Institute). Similarly, the CO
214 amounts derived from the Measurement of Pollution in the Troposphere (MOPITT), flying
215 aboard the NASA EOS-Terra satellite, are compared with WRF-Chem. MOPITT measures
216 the thermal infrared (IR) radiation with a spatial resolution of about $22 \text{ km} \times 22 \text{ km}$; these
217 radiances are then used to retrieve CO mixing ratios profile and total column amounts (Deeter
218 et al., 2003). Here we used version 6 Level 3 MOPITT CO data from the thermal infrared
219 band (TIR) to evaluate the spatio-temporal patterns of total column content simulated by the
220 model.

221 Tropospheric O_3 distributions simulated by WRF-Chem were compared to those derived from
222 the IASI-GOME2 multispectral approach, combining Infrared Atmospheric Sounding
223 Interferometer observations in the IR and Global Ozone Monitoring Experiment-2
224 measurements in the Ultraviolet (Cuesta et al., 2013, 2018). IASI-GOME2 has allowed the
225 first satellite observation of the horizontal distribution of O_3 pollution plumes located below 3
226 km of altitude, and also quantified the photochemical production of lowermost tropospheric
227 (LMT) O_3 across East Asia. This method is based on measurements from two instruments
228 onboard the MetOp satellite series since 2006 that offer global coverage every day with a
229 relatively fine ground resolution ($12 \text{ km} \times 25 \text{ km}$ for IASI at nadir and $80 \text{ km} \times 40 \text{ km}$ for
230 GOME-2). In this analysis, we have evaluated the LMT O_3 content integrating between the
231 surface and 3km height.

232 For the evaluation of aerosols simulations, we used aerosol optical depth (AOD) data at a
233 wavelength of 550 nm derived from the MODIS instruments onboard the Terra and Aqua
234 satellites. Level-2 MODIS Collection-6 data have a resolution of $10 \times 10 \text{ km}$ (at nadir). In this
235 analysis, we used the extinction coefficients simulated by WRF-Chem and extracted daily at
236 the satellite overpass time.

237 In our evaluation, we accounted for the satellite retrieval sensitivity by smoothing WRF
 238 vertical profiles with satellite averaging kernels (Kumar et al., 2012a). In the case of O₃, the
 239 IASI+GOME2 averaging kernel $A_{IASI+GOME2}$ and *a priori* O₃ profiles $X_{apriori}$ used by
 240 IASI+GOME2 were then applied to the WRF-Chem O₃ profile X_{int} (interpolated over the
 241 IASI+GOME2 vertical grid) to obtain smoothed WRF-Chem O₃ profile accounting for the
 242 satellite retrieval sensitivity, as classically done with the equation:

$$243 \quad X_{WRF-Chem(O_3)}^{smoothed} = X_{apriori} + A_{IASI+GOME2} [X_{int} - X_{apriori}] \quad (1)$$

244 A similar procedure was used to transform the modeled CO profiles using MOPITT averaging
 245 kernels and *a priori* profiles (Kumar et al., 2012a). For transforming the WRF-Chem
 246 simulated tropospheric NO₂ content for comparison to OMI retrievals, the procedure requires
 247 the user to calculate the transformed model profile (Y_{trop}) as:

$$248 \quad Y_{trop} = A \times \frac{AMF}{AMF_{trop}} \times X_{trop} \quad (2)$$

249 where A is the total column averaging kernel, AMF and AMF_{trop} are the air mass factors for
 250 the total columns and tropospheric columns, respectively, and X_{trop} is the tropospheric vertical
 251 profiles of NO₂, simulated by WRF-Chem, interpolated to the OMI pressure grid (Kumar et
 252 al., 2012a).

253 Finally, hourly NO₂, O₃, PM_{2.5} and PM₁₀ in-situ measurements were collected from 1497 air
 254 quality monitoring stations across China (589 rural and 908 urban), after checking for data
 255 quality. A minimum data capture of 75% was imposed to calculate seasonal mean
 256 concentrations.

257 2.3. Assessment of model performance

258 The model performance was evaluated over different seasons (January-February-March, JFM;
 259 April-May-June, AMJ; July-August-September, JAS; October-November-December, OND)
 260 by using the Pearson's correlation coefficient (r), mean bias (MB) and the fractional bias
 261 (FB). The first metric allows estimating the correlation pattern, thus the spatial agreement
 262 between model and observations. For physical parameters, the MB provides the absolute bias
 263 of the model, with negative and positive values indicating respectively underestimation and
 264 overestimation by the model while the FB (in %) is used for the chemical variables, as in this
 265 case the absolute bias would be hard to interpret. The mean biases were computed pointwise
 266 and then averaged over the whole domain:

$$267 \quad MB^j = \frac{1}{N_{obs}^j} \sum_{i=1}^{N_{obs}^j} (Mod_i^j - Obs_i^j) \quad (3)$$

$$268 \quad FB^j = \frac{1}{N_{obs}^j} \left[\frac{\sum_{i=1}^{N_{obs}^j} (Mod_i^j - Obs_i^j)}{\sum_{i=1}^{N_{obs}^j} \left(\frac{Mod_i^j + Obs_i^j}{2} \right)} \right] \times 100 \quad (4)$$

269 with Obs_i^j and Mod_i^j the observed and modeled values and N_{obs}^j the number of data at time i
 270 and station j over the domain. These metrics were successfully used in several studies for
 271 evaluating the performance of regional air quality models (e.g. Savage et al., 2013; Pope et
 272 al., 2015; Im et al., 2015; Crippa et al., 2016; Ghim et al., 2017; Crippa et al., 2019).

273 In case of in-situ data, we extracted WRF-Chem results at the lowest model layer and, for
 274 each station, we calculate the Pearson's correlation coefficient to assess the ability of the

275 model to reproduce the mean annual cycle and the mean bias to provide a measure model's
276 error; in the following analysis results are presented as an average over all the stations.
277

278 3. Results

279 3.1. Evaluation of meteorological variables

280 The general ability of the WRF model to reproduce realistic spatio-temporal patterns of most
281 relevant physical and chemical variables is assessed by comparing simulated output with
282 observational data averaged over different seasons for the reference year 2015. Looking at the
283 surface air temperature, WRF well captures the observed spatial pattern with a decreasing
284 south-north gradient and a cold area over the Tibetan plateau. In general, the spatial
285 distribution of 2-m temperature during all the seasons was very similar to ERA5 and only
286 slightly different from CRU (Fig. 1). The largest bias was observed during JFM, where the
287 model was warmer than the reference data over the northern part of the domain and colder in
288 most of western China, Vietnam, Laos, and Thailand (Fig. 1). In addition, large discrepancies
289 were observed over the Tibetan plateau, where the coarse resolution of the datasets and the
290 sparse availability of local measurements could explain the mismatch. The large thermal
291 heating occurring over the Indian region during spring and early summer (AMJ) is well
292 captured by the model, which is a pre-requisite to correctly simulate the summer Asian
293 monsoon. The high agreement in the spatial pattern is confirmed by the high spatial
294 correlation ranging between 0.98 and 0.99 with respect to both ERA5 and CRU datasets,
295 while the MB ranged from -1.51°C in winter (JFM) to 0.07°C in summer (JAS) using ERA5
296 as reference, and from -1.82°C in winter to -0.20°C in summer when compared to CRU
297 (Tab. 2).

298
299 Unlike seasonal variations in surface air temperature, which are mainly determined by the
300 insolation patterns, seasonal precipitation variations are strongly influenced by vertical
301 movement of air due to atmospheric instabilities of various kinds and by the flow of air over
302 orographic features; thus, to simulate accurately the seasonally varying pattern of
303 precipitation, models must correctly simulate a number of processes (e.g., evapotranspiration,
304 condensation, and transport) (Randal et al., 2007). The spatial distribution of seasonal mean
305 precipitation from model and reference data (ERA5 and CRU) is depicted in Fig 2. In general,
306 the model was able to capture the major convective centers as in the observations. WRF
307 overestimated the precipitation over the Himalayan region in JFM and over eastern China in
308 AMJ, nevertheless, it is important to note that WRF resolved the finer details of orographic
309 precipitation along the Himalayan foothills, which were missing both in ERA5 because of its
310 coarser resolution and in CRU due to the lack of widespread measurement stations. Besides,
311 WRF well reproduced the main monsoon features. The relevant thermal heating occurring
312 during AMJ over the Indian peninsula results in a pressure gradient with lows over the
313 landmasses and highs over the colder ocean, which causes a strong moisture advection from
314 the Bay of Bengal to Indian Peninsula leading to increased precipitation over South-East Asia
315 and the Indian landmass region during JAS. Looking at the spatial agreement, except for CRU
316 in winter ($r = 0.73$), the correlation coefficient exceeded 0.80 during all the seasons and for
317 both the datasets. The area averaged bias, with respect to the CRU observation, was 0.21-0.25
318 mm/day during the cold periods (OND-JFM) and 0.68-0.74 mm/day during the warm periods
319 (AMJ-JAS), while compared to ERA5, WRF showed a slight wet bias during the warm
320 periods (+ 0.15-0.27 mm/day) and a slight dry bias (about -0.05 mm/day) during the cold
321 seasons (Tab. 2). Large discrepancies (overestimation) were observed between 2-m relative
322 humidity and CRU observations over central Asia, Western China and India (Fig. 3) while an

323 underestimation was observed over the western part of the domain compared to ERA5 during
324 JAS and OND. The spatial correlation ranged from 0.95 to 0.99 for both ERA5 and CRU
325 datasets. The WRF model underestimated the relative humidity with respect to ERA5 during
326 all the year, from - 1.5% in JFM to - 4.5% in JAS, and an overestimation was observed
327 compared to CRU dataset, in particular during the warm period with a MB of 11-12%.

328
329 Looking at latent heat we found an overall good agreement between the WRF model and the
330 reference datasets (Fig. 4), with spatial correlation exceeding 0.92 for both datasets (Tab. 2).
331 Nevertheless, WRF remarkably overestimated surface latent heat during the warm seasons,
332 especially over South and Southeastern Asia, with the largest bias found in JAS and ranging
333 between 2.6 W/m² and 13.2 W/m² compared to ERA5 and MTE, respectively. This
334 overestimation was strictly related with the slight overestimation of rainfall during the same
335 season which brings more water on the land surface that can re-evaporate, subsequently
336 amplifying convective precipitation. Similarly, the sensible heat was well simulated with a
337 spatial correlation ranging from 0.82 in JFM for MTE dataset to 0.94 in AMJ for ERA5.
338 Compared to ERA5, WRF model overestimated the sensible heat during the warm period
339 (16.4-19.4 W/m²), in particular over the Western part of the domain, while a lower
340 overestimation is reported for MTE (7.9-11.5 W/m²) over the same season. In contrast during
341 JFM and OND, WRF model well reproduced the observed spatial patterns (Fig. 5) with a
342 slight underestimation in winter (- 1.2 and - 2.8 W/m²).

343
344 Besides, WRF well reproduced the snow cover fraction, with a spatial agreement ranging
345 from 0.74-0.77 in summer to 0.92-0.95 in winter for ERA5 and MODIS datasets, respectively
346 (Tab. 2). Compared to MODIS data, WRF slightly overestimated the snow cover fraction in
347 JFM (0.6%) and AMJ (0.2%) mainly around the Tibetan plateau and underestimated it in JAS
348 (about - 0.1%) and OND (- 1.7%), WRF underestimated the snow cover throughout the year
349 when compared to ERA5, with a mean bias ranging from - 0.9% in JAS to - 3.2% in AMJ.
350 The main discrepancies were observed over North Siberia in JFM, over the Tibetan plateau in
351 AMJ and over both areas in OND (Fig. 6).

352

353 **3.2. Evaluation of chemical variables**

354 The spatial distributions of model-simulated and OMI-retrieved seasonal mean tropospheric
355 NO₂ content during winter, spring, summer and autumn for the year 2015 are shown in Fig. 7.
356 Both WRF-Chem and OMI showed, during all the seasons, the highest tropospheric NO₂
357 content over the polluted region around Beijing and over Korean peninsula, followed by the
358 Indo-Gangetic Plain region, with hot spots located in correspondence of large urbanized areas
359 such as Seoul (South Korea) and New Delhi (India). The lowest values were found above the
360 Tibetan plateau. The spatial correlation averaged over the entire domain ranged between a
361 0.89 in AMJ and 0.91 in JFM. The percentage differences between WRF-Chem and OMI
362 tropospheric NO₂ content showed a slight underestimation of the model by 2-8% during the
363 cold period (Tab. 3) while the concentrations of NO₂ were significantly under-predicted by
364 WRF-Chem during the warm period (64-70% in AMJ-JAS). Conversely, large
365 overestimations occurred during the cold months (JFM and OND), mostly over polluted
366 regions (e.g. Eastern China). The positive and negative biases found in summer and winter,
367 respectively, compensated with each other and led to an overall small FB (- 2%).

368 The highest total column CO was observed over the Southeastern and Eastern Asia and the
369 lowest column CO values were found above the Tibetan plateau (Fig. 8). The simulated total
370 CO column showed a high spatial correlation coefficient (about 0.97) during all the seasons
371 (Tab. 3). The model performed well for simulating CO with a FB < ± 10% of MOPITT. The

372 agreement between WRF-Chem and MOPITT is higher in autumn (FB: + 3%) than in spring
373 (FB: + 7%) and summer (FB: + 8%). The simulated total column CO was slightly
374 underestimated over Eastern Asia during all seasons and over Southeastern Asia in winter and
375 autumn. In summer, the most important overestimation was observed over Southeastern Asia
376 and India (Fig. 8).

377 The spatial and seasonal distributions of model-simulated and IASI+GOME2-retrieved LMT
378 O₃ content (integrated up to 3km height) are shown in Fig. 9. The WRF-Chem model
379 simulated well the spatial distributions and the seasonal variations of O₃. A particular good
380 spatial agreement is remarked for the summer season, with a spatial correlation of 0.99 (Tab.
381 3). During spring and winter, the spatial agreement is moderate ($r = 0.61$ and $r = 0.76$,
382 respectively), while it is fairly low in winter ($r = 0.43$). The highest O₃ content was similarly
383 depicted in Eastern Asia and in the latitude band 20-45°N, in particular over the Indo-
384 Gangetic Plain region, in both datasets. Although overestimated in the cold seasons, WRF-
385 Chem simulates correctly the eastward export of tropospheric O₃ over the Yellow and Japan
386 Sea (30-40°N 120-130°E). Model and satellite data also agree to show very low LMT O₃
387 values above the Tibetan plateau during the cold season and below 30°N latitude in summer.
388 North of these high mountains (40°N 80-100°E), high abundances of O₃ during summer and
389 spring depicted by IASI+GOME2 are consistently simulated by WRF-Chem. The overall
390 seasonal cycle is consistently shown by both datasets, with highest concentrations in spring, a
391 little lower in summer and lowest in autumn. The FB over the model domain exhibited a
392 seasonal variability with limited overestimation during the warm season (3-7% in spring and
393 summer), in the 20-30°N latitude band, and larger over-prediction (11-13%) during the cold
394 period (winter and autumn). In addition, we remark that only IASI+GOME2 retrievals show
395 moderate enhancement of O₃ concentrations north of 45°N during winter, probably associated
396 with downward transport of O₃ from both upper troposphere and stratosphere. This difference
397 between IASI+GOME2 and model data was also remarked in a comparison with respect to
398 other simulations performed with WRF-Chem in springtime 2009 over East Asia (Cuesta et
399 al., 2018).

400 Looking at the aerosol optical depth (Fig. 10), both WRF-Chem and MODIS showed, during
401 all the seasons, the highest AOD over the polluted region of Eastern Asia, over the Goby
402 desert and over the Indo-Gangetic Plain region. The spatial correlation ranged between 0.86 in
403 summer and 0.93 in winter (Tab. 3). The model-simulated AOD were lower than those from
404 MODIS with a FB of - 19% and - 2% for JFM and OND, respectively, in particular over the
405 polluted region of Eastern China and Indo-Gangetic Plain region (Fig. 10). WRF-Chem
406 overestimated the AOD by 6% and 20% in spring and summer over Southern part of the
407 domain.

408 Compared to in-situ measurements, WRF-Chem was able to reproduce the surface NO₂
409 concentrations over China during all the seasons (Fig 11). The high concentrations over
410 polluted regions are well captured, despite some stations showed a large bias, while minimum
411 NO₂ concentrations, found in Western China, are slightly underestimated. Overall, the
412 correlation coefficient computed from mean daily concentrations was 0.29, with a mean bias
413 of 10.1 ppb and a FB ranging from 35% in spring to 45% in autumn (Tab. 4). These statistics
414 were calculated from the mean of the metrics computed over the stations, thus stations with
415 poor agreement significantly contribute to lower model skills. As poor model performances
416 were expected, particularly in urban areas, where a regional chemistry transport model is
417 unable to correctly predict the observed hourly variability of air pollutants concentrations,
418 which depends on local processes, we also computed the correlation comparing the mean
419 temporal evolutions averaged over all the stations. In this latter case, the model performance

420 was remarkably better, with a temporal correlation of 0.71 (data not shown). Unlike NO_2 , the
421 comparison of surface O_3 showed a complex bias pattern: the model well reproduced O_3
422 during cold seasons (FB = - 8% in JFM, - 15% in OND), in particular concentrations below
423 15 ppb over megacities (Fig. 12). Similarly, high O_3 concentrations over Tibetan plateau are
424 well represented during all the seasons. Nevertheless, during warmer months (AMJ and JAS),
425 the WRF-Chem systematically overestimated surface O_3 concentrations (FB = 23-24%). The
426 mean annual cycle of O_3 is slightly better simulated compared to NO_2 , with a correlation
427 coefficient of 0.51 (0.90 in case of correlation computed from the mean temporal evolution
428 averaged over all the stations), while the mean bias was 5 ppb (Tab. 4). Looking at surface
429 $\text{PM}_{2.5}$ (Fig. 13) and PM_{10} (Fig. 14) concentrations during the cold period, the high
430 concentrations (exceeding $100 \mu\text{g m}^{-3}$) over polluted regions and megacities in Eastern China
431 are well captured (FB = 5% in JFM for both $\text{PM}_{2.5}$ and PM_{10}), while low $\text{PM}_{2.5}$ and PM_{10}
432 concentrations in Western China (less than $10 \mu\text{g m}^{-3}$) are slightly overestimated. During the
433 warm period, the surface concentrations are well captured across China, with a slight
434 overestimation in AMJ for PM_{10} (FB = 11%). The highest overestimation is observed in
435 summer with a FB of 29% and 36% for PM_{10} and $\text{PM}_{2.5}$, respectively (Tab. 4). Overall, the
436 correlation coefficient computed from mean daily concentrations was 0.44 for $\text{PM}_{2.5}$ and 0.35
437 for PM_{10} , with a mean bias of $10.4 \mu\text{g m}^{-3}$ and $14.8 \mu\text{g m}^{-3}$, respectively (Tab. 4). By
438 considering the mean temporal evolutions averaged over all the stations, the temporal
439 correlations were 0.83 and 0.71, respectively (data not shown).

440

441 4. Discussion

442 Capturing spatiotemporal patterns of trace gases and weather patterns over Asia is challenging
443 for chemistry transport models because of the complex orography associated to the monsoon
444 systems and large uncertainty in the anthropogenic emission inventories over heavily
445 populated regions in Asia (Kumar et al., 2015; Zhang et al., 2016a). In this study, the WRF-
446 Chem model was used to simulate the spatial and seasonal variability of main physical and
447 chemical variables over the Asian region at fine horizontal resolution (8 km) to capture local
448 small-scale processes.

449 In addition to surface air temperature and moist fluxes, which influence the rate of chemical
450 reactions close to land surface and the removal of air pollutants through wet deposition,
451 respectively, heat fluxes also play a pivotal role in surface energy balance and influence the
452 Asian monsoon (Wang et al., 2014, 2016, 2017b). When compared to ERA5 and CRU
453 datasets, WRF well reproduced meteorological observational-based data. In particular,
454 looking at temperature, WRF performed well in terms of spatial distributions over time, even
455 over complex terrain with uneven surface topography, such as Tibetan plateau and Himalayan
456 chain. To reproduce observed patterns over Tibetan plateau, a meteorological model must
457 correctly reproduce several processes ranging from convection to thermal balance.
458 Considering the relative humidity, we found slight discrepancies over the Tibetan plateau,
459 particularly in winter, while we found a large bias for surface sensible heat over the Western
460 part of the domain during the warm period. Besides, WRF slightly overestimated the
461 precipitation, in particular during the monsoon period; previous studies suggested that the
462 magnitude of precipitation bias depends on the cumulus parameterization schemes (Ratna et
463 al., 2014; Juneng et al., 2016). The WRF model predicted much stronger rain over Western
464 China, Northern Bay of Bengal and Eastern India. Zhang et al. (2016a) showed that the
465 rainfall overestimation can be attributed to the slightly excessive precipitation predicted by
466 the cumulus Tiedtke scheme, and the apparent underestimation of air temperature is most
467 likely caused by an error in the radiation balance and certain limitations in the Yonsei

468 University Scheme used in resolving main features of the Planetary Boundary Layer
469 meteorology, particularly over complex terrain such as mountainous regions over high-
470 elevation Tibetan plateau, Vietnam and Laos (Zhang et al., 2016b).

471 A previous study performed with WRF over South Asia at 45 km of spatial resolution,
472 indicated that the MB was 1-4 °K for temperature, 20-65 % for water vapor and within ± 10
473 mm/day for the precipitation during all seasons except in summer, with an overestimation
474 exceeding 20 mm/day over Himalaya and along the coastline in Eastern India (Kumar et al.,
475 2012b). In another study where WRF was run over East Asia at 36 km of horizontal
476 resolution, Zhang et al. (2016b) showed a MB ranging from - 1.0°C to + 1.5°C for surface air
477 temperature and moderate to large biases for precipitation (+ 0.2 to + 1.7 mm/day) and
478 relative humidity (+ 0.4% to + 23.4%). In a different study over East Asia, where WRF was
479 run at 50 km of horizontal resolution, using a spectral nudging applied to wind direction and
480 speed and air temperature over the time period 1989-2007, Tang et al. (2017) found for the
481 surface air temperature an averaged MB of 1.77 °C (- 8 to + 4 °C) compared to CRU, and of
482 1.45 mm (- 4 to + 8mm) for precipitation with an overestimation by 2-8 mm/day in tropical
483 regions and an underestimation of 0.5-1.0 mm/day over Southern China.

484 In our study, the model performances in simulating surface meteorology were better than the
485 other simulations performed over Asian regions with the same model but with a coarser
486 spatial resolution at 36 - 50 km of spatial resolution (e.g. Kumar et al., 2012a,b; Zhang et al.,
487 2016a,b; Tang et al., 2017); this suggests that the finer model resolution (8 km) helps to
488 resolve small-scale features induced by complex topography e.g. in Himalaya or Sichuan
489 region. In general, increasing model resolution has resulted in improved model simulations
490 and predictions for air temperature, relative humidity, precipitation (Malardel et al., 2016;
491 Prodhomme et al., 2016; Zhang et al., 2016c), global energy budget (Vannière et al., 2019)
492 and orographic winds (Roebber et al., 2004). By using the WRF model with horizontal
493 resolutions of 2, 10 and 30 Km, Lin et al. (2018) showed that finer resolutions improved
494 biases over the Tibetan Plateau, in particular for precipitation. A significant difference was
495 observed from 30 to 10 km of horizontal resolution, suggesting that approximately 10 km of
496 horizontal resolution represents a good compromise (Lin et al., 2018).

497 The WRF-Chem model well reproduced tropospheric NO₂ content, total column CO and LMT
498 O₃ content with a FB within the air quality model performance criteria, except for NO₂ during
499 the warm season. The highest column content for NO₂, CO and O₃ was observed over
500 Southeastern and Eastern Asia due to road traffic, industries, power plants and biomass
501 burning (Streets et al., 2003; Kumar et al., 2012a; Cooper et al., 2014), with 2015 recording
502 the highest fire activity season since 1997 (Huijnen et al., 2016; Mead et al., 2018). In
503 contrast, the column NO₂, CO and O₃ values above regions of high terrain such as the Tibetan
504 plateau were lower than adjacent regions due to the limited depth of the troposphere and
505 fewer emissions (Cooper et al., 2014).

506 In this study, WRF-Chem captured the seasonal variability of tropospheric NO₂ content
507 values with a summer minimum and a winter maximum. The winter maximum is due to a
508 lower removal rate of NO₂ with OH radicals, compared to summer (Beirle et al., 2003). The
509 large NO₂ overestimation was observed during winter and autumn over polluted regions with
510 abundant anthropogenic NO_x emissions, such as road traffic and power plants as previously
511 reported e.g. in Europe (Barten et al., 2019; Visser et al., 2019). These results are in
512 agreement with prior studies that found WRF-Chem overestimating tropospheric NO₂ content
513 in urban areas such as London, Madrid, Rome and in cities of Eastern Europe by 5-18 % in
514 urban areas (Barten et al., 2019; Visser et al., 2019), despite they adopted a different chemical

515 mechanism (i.e., CBM-Z). The NO_x emissions from microbial activity and lightning, which
516 were not considered in our emission inventory, are important during summer in rural areas of
517 Southeastern Asia (Kumar et al., 2012a). The largest discrepancies (underestimation) during
518 spring and summer point to uncertainties in biomass burning, lightning and soil emission
519 estimates (Kumar et al., 2012a; Barten et al., 2019) and another plausible reason is the NO_x
520 removal overestimation through the reaction of dinitrogen pentoxide (N_2O_5) to nitric acid in
521 the WRF-Chem chemical mechanism (Yegorova et al., 2011). The discrepancies in NO_x
522 concentrations can be also explained by no year adjustments for EDGAR-HTAP
523 anthropogenic emission data (based on year 2010), in particular in China. Kumar et al.
524 (2012a) showed that the discrepancies in anthropogenic emission estimates were mainly due
525 to uncertainties in the emissions inventory (emission factors and socio-economic parameters).
526 Prior studies have found that, over South Asia, WRF-Chem tends to overestimate NO_2
527 tropospheric content from OMI retrievals by 10-50% over South Asia and up to 90% over the
528 Indo-Gangetic Plain region during winter (Kumar et al., 2012a) with a correlation coefficient
529 between model and OMI ranging from 0.61 to 0.73 in 2008 (Kumar et al., 2012a). Over East
530 Asia, WRF-Chem underestimated the tropospheric NO_2 content by up to - 30.6% compared to
531 SCIAMACHY data in 2005 (Zhang et al., 2016b). As NO_x emissions display strong spatial
532 variation, we obtained a better spatial representation and simulations of NO_2 levels, by
533 increasing the model grid resolution, compared to previous studies at coarser scale e.g. at
534 20 km of grid resolution (Schaap et al., 2015; Barten et al., 2019; Visser et al., 2019).
535

536 The seasonal variation of the total column CO is well reproduced by WRF-Chem with highest
537 and lowest values during late autumn-winter and summer (monsoon), respectively. In general,
538 both the model and MOPITT were highest during winter, decreased during spring, attained
539 minimum levels during summer and increased again during autumn. MOPITT CO retrievals
540 over South and Southeast Asia were slightly underestimated by WRF-Chem in spring,
541 between March and May, when biomass burning constitutes the major fraction of total CO
542 emissions (Amnuaylojaroen et al., 2019), suggesting that CO emissions from biomass burning
543 is slightly underestimated. Over South Asia, the WRF-Chem model similarly estimated
544 MOPITT column CO retrievals by - 9.0% to + 7.0% during all seasons with a r value from
545 0.63 to 0.84 for the year 2008 (Kumar et al., 2012a) and by - 24.2% to + 3.9% over East Asia
546 in 2005 (Zhang et al., 2016b). The annual mean contribution of biomass burning to the total
547 CO emissions was around 24% over Asia (Streets et al., 2003). The slight overestimation of
548 CO retrievals during other seasons (low fire activity) indicated that anthropogenic CO
549 emissions are overestimated over this region (Zhang et al., 2016a). A better treatment of
550 biomass burning sources and improved boundary conditions of CO (e.g. for transboundary
551 inputs, in particular from wildfires, biomass burning and transport) are needed to improve the
552 performance of the total column CO.
553

554 The seasonality of LMT O_3 content is well reproduced by WRF-Chem by capturing the
555 increase in O_3 burden during the warm season, with a spring maximum (Sicard et al., 2009;
556 Kumar et al., 2012a; Cooper et al., 2014). Eastern China and Northern India are two main
557 pollution sources, emitting significant amounts of NO_x , CO and VOCs (Wang et al., 2010) in
558 winter and autumn (cold period) leading to the highest LMT O_3 in spring and summer
559 (Cooper et al., 2014). Furthermore, the relatively high biogenic NMVOC emissions and active
560 photochemical reactions constitute favorable conditions for O_3 formation in summer (Sicard
561 et al., 2016b). The WRF-Chem overestimation of anthropogenic NO_2 and CO emissions led to
562 a model overestimation of surface O_3 concentrations in winter (13%) and autumn (11%), in
563 particular in South Asia and Eastern China. A previous study employing an offline regional
564 model showed an overestimation of O_3 levels during summer over India (Roy et al., 2008). As

565 reported here, the regional models generally underestimate mean O₃ concentrations during
566 high O₃ seasons and overestimate mean O₃ concentrations during low O₃ seasons (Fiore et al.,
567 2009; Huang et al., 2017). The differences in anthropogenic NO_x, CO and VOCs inventories
568 are the dominant factors for the discrepancies in simulated O₃ levels, as already reported in
569 China and Southeast Asia (Ma and van Aardenne, 2004; Amnuaylojaroen et al., 2014). The
570 slight mean bias of LMT O₃ for spring 2015 (3%) entails among other the correct
571 representation of the seasonal variations of stratospheric O₃ intrusions to the upper
572 troposphere. The over-prediction of O₃ and under-prediction of NO₂ in all months indicated
573 an insufficient titration of O₃ by NO (Zhang et al., 2016b). In Southern India and Southeastern
574 Asia, below 30°N latitude, the summer is dominated by cloudy conditions and heavy rainfall
575 due to monsoon leading to lower O₃ levels by reducing the photochemical production of O₃
576 (e.g. Roy et al., 2008; Kumar et al., 2012a). The FB ranged from - 16% to 0% over South
577 Asia in 2008 (Kumar et al., 2012a) with larger differences during spring and early summer,
578 mainly due to additional O₃ precursor sources (e.g. biomass burning).

579
580 High AOD were observed over desert regions due to mineral dust (e.g. Goby) and over areas
581 with large anthropogenic aerosol emissions, especially East and South Asia (Shindell et al.,
582 2013). As EDGAR does not provide black and organic carbon and PM_{2.5} emissions, and
583 GOCART does not include secondary organic aerosols or nitrate aerosols, an under-prediction
584 of aerosol burden is observed in Asia, in particular in winter (Zhong et al., 2016; Zhang et al.,
585 2016a; Crippa et al., 2019). The overestimations of AOD over Eastern China, South and
586 Southeastern Asia in spring and summer, were mainly due to over-predictions in PM₁₀
587 concentrations because of dust emissions (Shindell et al., 2013; Zhang et al., 2016a). Over
588 East Asia, the differences between model-simulated and MODIS-based AOD ranged from -
589 38.7% to + 5.6% in 2005 (Zhang et al., 2016b).

590 Considering the performances of WRF-Chem for reproducing atmospheric chemistry at
591 ground-level, the recommended benchmarks proposed by Morris et al. (2005), Boylan and
592 Russell (2006) and Emery et al. (2017) for the chemical model performance is FB within \pm
593 15% for O₃ and NO₂ if r is greater than 0.5. By comparing the WRF-Chem model outputs
594 with ground-based observations, overall the WRF-Chem model reproduced well the mean
595 annual cycle of surface O₃, PM_{2.5} and PM₁₀ mean concentrations at regional scale in 2015, in
596 particular during the cold season. Furthermore, the model well reproduced the NO₂ titration
597 over the polluted Eastern China. The WRF-Chem model overestimated surface O₃
598 concentrations during the warm period, when plants are active, then a part of the observed
599 bias can be explained by (i) a poor parameterization of dry deposition to vegetation in the
600 Noah Land Surface Model, leading to an underestimation of dry deposition velocities (Wu et
601 al., 2011), then to a reduction of the O₃ removal capacity by plants; and by (ii) the under-
602 prediction of cloud optical depth and overestimation of photolysis rates by WRF (Ryu et al.,
603 2018). However, when the correlations were computed from the average of individual
604 stations, the performances were much poorer. Nevertheless, this result was partially expected
605 being an intrinsic characteristic of regional models. In other words, a regional CTM (even at
606 high spatial resolution) is not able to reproduce well temporal variation of local urban
607 observations because of the lack of detailed local inputs (both emission inventories and
608 removal processes). Besides, it should be noted that WRF-Chem was forced with annual mean
609 anthropogenic emissions, thus it is hard to reproduce the high frequency temporal variability
610 of observations, especially in urban environments which are extremely dynamics and poorly
611 controlled by natural processes.

612

613 5. Conclusions

614 In this study WRF-Chem was applied at high spatial resolution (8-km) over Asia for the year
615 2015, to allow resolving fine-scale features over complex topography areas (e.g. Indo-
616 Gangetic Plain region). As we did not use nudging, our results enhance the reliability of this
617 study and the ability of the model to capture spatio-temporal variation of physical and
618 chemical variables. Overall model skills in simulating surface meteorological and air quality
619 were higher than those showed in previous studies focusing on the same region where the
620 model was applied at coarser spatial resolution. In this study, the mean bias ranges are lower
621 than Zhang et al. (2016b) e.g. [- 1.51 °C; + 0.07 °C] vs. [- 1.0 °C; + 1.5°C] for air
622 temperature, [- 0.05 mm/day; + 0.27 mm/day] vs. [+ 0.2 mm/day; + 1.7 mm/day] for
623 precipitation; [- 4.5 %; - 1.5 %] vs. [+ 0.4 %; + 23.4%] for relative humidity and [- 7 %; + 8
624 %] vs. [- 24.2 %; + 3.9 %] for column CO retrievals. The better observed performance is
625 mainly due to the advantage of the finer model spatial resolution. WRF-Chem simulation
626 showed low to moderate biases for major meteorological variables, except for sensible heat
627 where a large bias (16-19 W.m⁻²) is observed during the warm period. Furthermore, the results
628 indicate some limitations in parameterization, such as the cumulus Tiedtke scheme leading to
629 precipitation overestimation over widely-varying climate and topography regions e.g. high-
630 elevation Tibetan plateau, Indo-Gangetic Plain region and Southeastern Asia.

631
632 The WRF-Chem reproduced well the overall spatial and seasonal variability of O₃, CO and
633 AOD over the Asian region, but large discrepancies were found for NO₂ during the warm
634 period. The observed bias between model-simulated and satellite-retrieved values were
635 mainly attributed to uncertainties in satellite retrievals, vertical and horizontal model
636 resolution, bottom-up emissions, anthropogenic and natural NO_x emission estimates (e.g. by
637 lightning and soil), dust and PM_{2.5} emission, stratosphere-to-troposphere O₃ exchange,
638 transboundary pollution, low NO titration and uncertainty in N₂O₅ and nitrate radical
639 reactions in the WRF-Chem chemical mechanism (Yegorova et al., 2011; Li et al., 2014;
640 Parrish et al., 2014; Huang et al., 2017; Sicard et al., 2017; Zhang et al., 2016a; Mu et al.,
641 2017).

642
643 For any application of models results, including reliable air pollution risk assessment, the
644 generation of realistic maps is needed, particularly over highly complex terrain of Northern
645 India (Indo-Gangetic Plain) where air quality is poor (Kumar et al., 2012a). Regional
646 chemistry-climate models at coarse horizontal resolution are often unable to resolve the local
647 features influencing the chemical transformation (Tie et al., 2010; Huang et al., 2017) and
648 barely able to fully reproduce the ground observations (Schaap et al., 2015; Jonson et al.,
649 2018) in particular at high-elevation sites (Strode et al., 2015). To date, the most damaging air
650 pollutant for vegetation and human health are O₃ and PM (Sicard et al., 2016a, 2019). In this
651 study, WRF-Chem model reproduced well the spatial and seasonal variability of surface NO₂,
652 O₃, PM_{2.5} and PM₁₀ mean concentrations across China, following the recommended
653 benchmarks for the chemical model performance for O₃ and NO₂. However, our results
654 suggest that it is essential to improve the emission estimates of primary PM (e.g. new dust
655 emission scheme) and NO_x (e.g. adjusted anthropogenic emission and vertical distribution)
656 and upgrade chemical mechanisms e.g. Polycyclic Aromatic Hydrocarbon heterogeneous
657 reactions with O₃ and homogeneous reaction with the nitrate radical in order to reduce bias in
658 simulating the surface O₃ and PM concentrations over Asia (Zhang et al., 2016b; Mu et al.,
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660
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676 **Data availability.** All data and figures are available in this paper. No more data are available.

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678

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1208 emissions and evaluation of simulated air quality”. *Geosci. Model Dev.*, 9: 1201-1218.
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Journal Pre-proof

1210 **Table 1.** Model set up with main physical and chemical schemes adopted in the simulation.

Model set-up	Values
Domain	South-East Asia
Simulation period	1 st January - 31 st December 2015
Spin up	20 st December - 31 st December 2014
Horizontal resolution	8 km
Vertical resolution	30 eta levels up to 50 hPa
Domain size	780 x 690 cells (lon x lat)
Meteorological boundary	ERA5 (31 km), 3h
Chemical boundary	MOZART-4/GEOS-5
Physical option	Adopted scheme
Microphysics	Single-moment 6-class
Cumulus Parameterization	New Tiedtke
Shortwave Radiation	RRTMG
Longwave Radiation	RRTMG
Land-surface	Noah Land Surface Model
Planetary boundary layer	Yonsei University Scheme
Chemical options	Adopted scheme
Gas phase chemistry	MOZART
Aerosols	GOCART
Photolysis	Madronich F-TUV
Biogenic emissions	MEGAN
Anthropogenic emissions	EDGAR HTAP (v2.2)
Fire emissions	FINN (v1.5)

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1214 **Table 2.** Pearson's correlation coefficient (r) and mean bias (model-reference) computed
 1215 using WRF results and different reference datasets for surface air temperature, precipitation,
 1216 relative humidity, latent heat, sensible heat and snow cover fraction. Note that reference data
 1217 were re-gridded to the WRF domain; in addition to downscale temperature we used a dry
 1218 adiabatic lapse rate correction.

VARIABLE	REFERENCE	Pearson's correlation coefficient (r)				Mean bias			
		<i>JFM</i>	<i>AMJ</i>	<i>JAS</i>	<i>OND</i>	<i>JFM</i>	<i>AMJ</i>	<i>JAS</i>	<i>OND</i>
TEMPERATURE (°C)	ERA5	0.99	0.99	0.99	0.99	-1.51	-0.42	0.07	-0.68
	CRU	0.98	0.99	0.99	0.99	-1.82	-0.82	-0.20	-1.20
PRECIPITATION (mm/day)	ERA5	0.80	0.90	0.91	0.88	-0.04	0.15	0.27	-0.05
	CRU	0.73	0.84	0.84	0.81	0.25	0.74	0.68	0.21
RELATIVE HUMIDITY (%)	ERA5	0.99	0.99	0.99	0.99	-1.5	-3.1	-4.5	-4.0
	CRU	0.95	0.96	0.97	0.96	2.1	11.0	12.0	2.7
LATENT HEAT (W/m ²)	ERA5	0.92	0.94	0.95	0.94	2.0	0.5	2.6	-0.24
	MTE	0.95	0.97	0.97	0.95	3.0	6.3	13.2	4.6
SENSIBLE HEAT (W/m ²)	ERA5	0.88	0.94	0.93	0.87	-1.2	16.4	19.4	2.2
	MTE	0.82	0.93	0.91	0.87	-2.8	7.9	11.5	1.5
SNOW COVER FRACTION (%)	ERA5	0.95	0.84	0.74	0.89	-3.0	-3.2	-0.9	-2.7
	MODIS	0.92	0.89	0.77	0.92	0.6	0.2	-0.08	-1.7

1219 *JFM: January-February-March, AMJ: April-May-June, JAS: July-August-September, OND: October-*
 1220 *November-December.*

1221

1222

1223 **Table 3.** Pearson's correlation coefficient (r) and fractional bias computed using WRF-Chem
 1224 results and different reference datasets for surface nitrogen dioxide (NO₂), aerosol optical
 1225 depth (AOD), ozone (O₃) and carbon monoxide (CO) concentrations.

VARIABLE	REFERENCE	Pearson's correlation coefficient (r)				Fractional bias			
		JFM	AMJ	JAS	OND	JFM	AMJ	JAS	OND
NO ₂	OMI	0.91	0.89	0.90	0.89	-2%	-64%	-70%	-8%
CO	MOPITT	0.97	0.97	0.96	0.97	-7%	7%	8%	3%
O ₃	IASI	0.76	0.61	0.99	0.43	13%	3%	7%	11%
AOD	MODIS	0.93	0.88	0.86	0.91	-19%	6%	20%	-2%

1226 *JFM: January-February-March, AMJ: April-May-June, JAS: July-August-September, OND: October-*
 1227 *November-December.*

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1230 **Table 4.** Pearson's correlation coefficient, mean bias and fractional bias computed using
 1231 WRF-Chem daily results at surface layer and ground measurements for surface nitrogen
 1232 dioxide (NO₂), ozone (O₃), particulate matters (PM_{2.5} and PM₁₀) concentrations; results
 1233 represent the mean computed over all the available stations.

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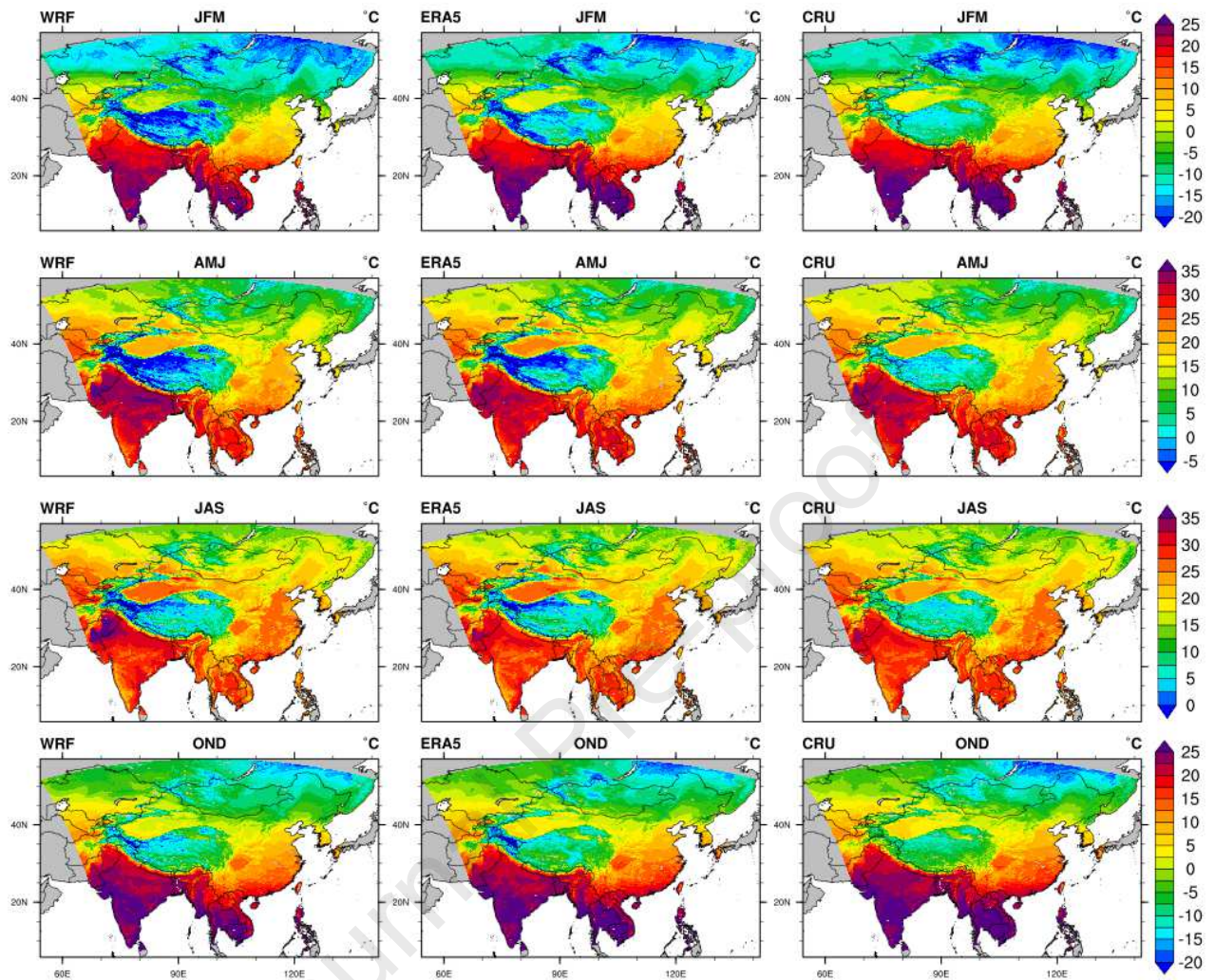
VARIABLE	Pearson's correlation coefficient (r)	Mean bias	Fractional bias			
			JFM	AMJ	JAS	OND
NO ₂	0.29	10.1 ppb	38%	35%	42%	45%
O ₃	0.51	5.0 ppb	-8%	23%	24%	-14%
PM _{2.5}	0.44	10.4 µg m ⁻³	5%	20%	36%	23%
PM ₁₀	0.35	14.8 µg m ⁻³	5%	11%	29%	24%

1235 *JFM: January-February-March, AMJ: April-May-June, JAS: July-August-September, OND:*
 1236 *October-November-December.*

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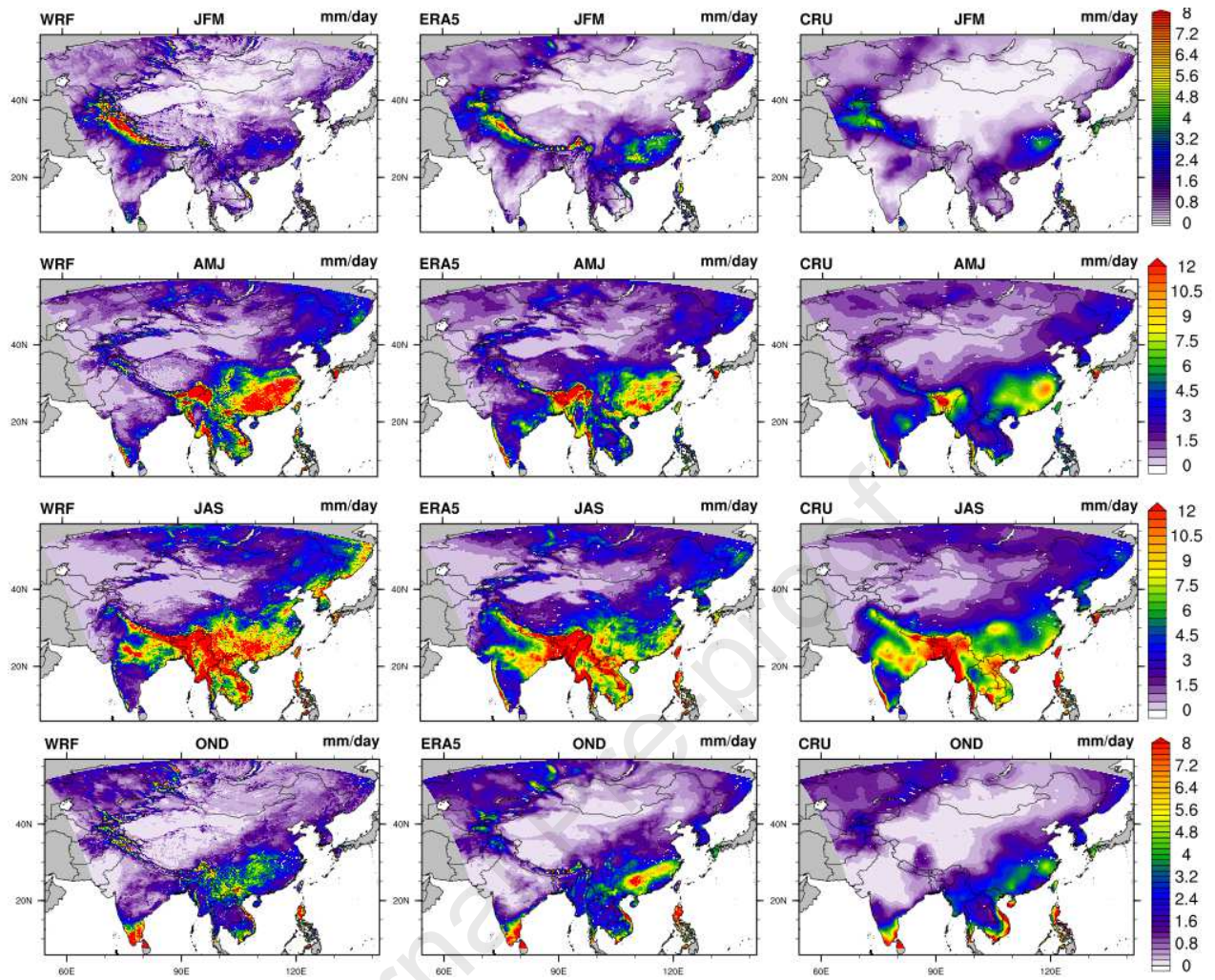
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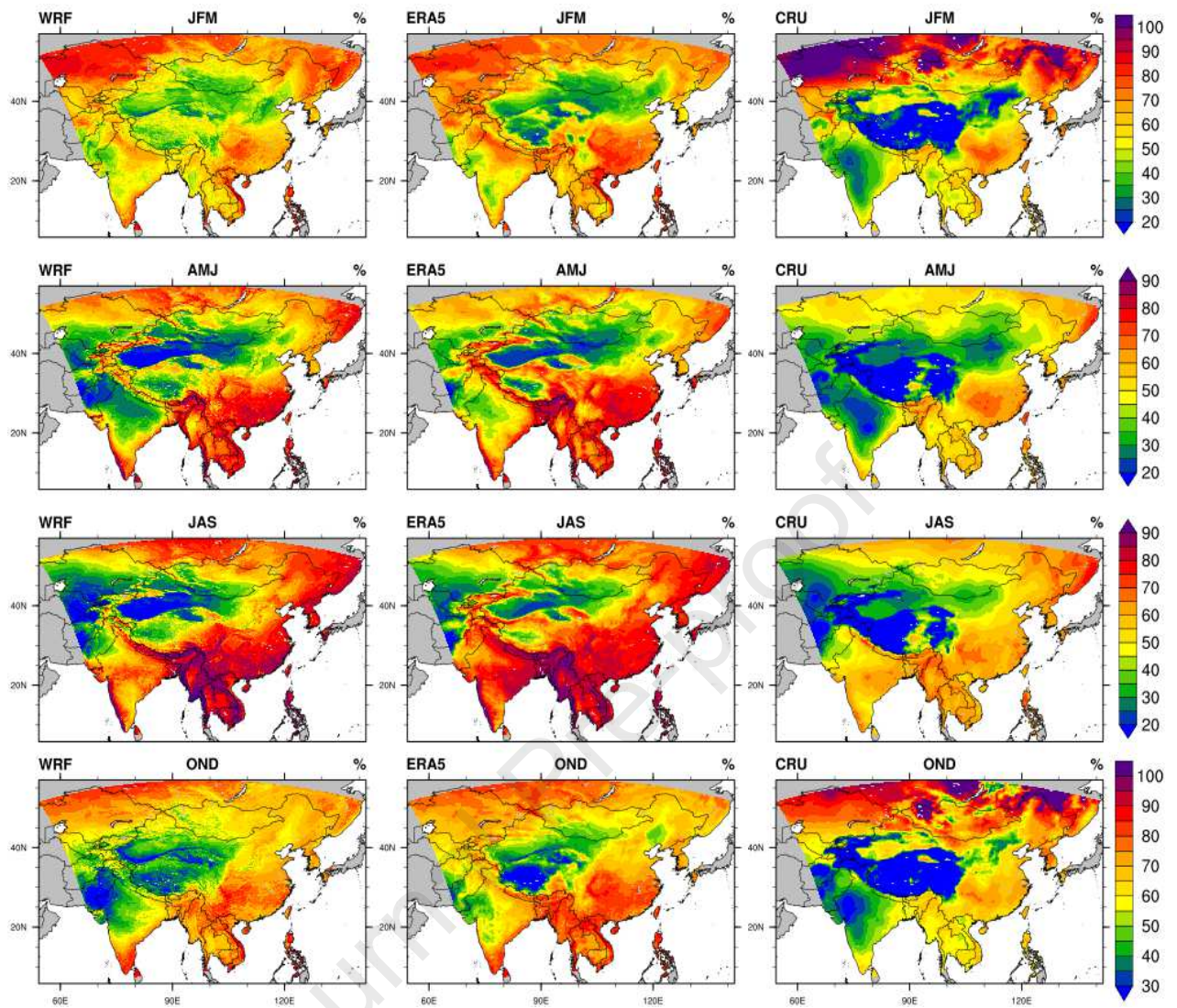
1241 **Figure 1.** Spatial pattern of 2-meter air temperature (°C) as simulated by WRF-Chem (left
 1242 panels) and compared to ERA5 reanalysis (central panels) and CRU dataset (right panels)
 1243 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-
 1244 August-September, OND: October-November-December) in 2015. Mind the differences in
 1245 color scales between seasonal climatologies.



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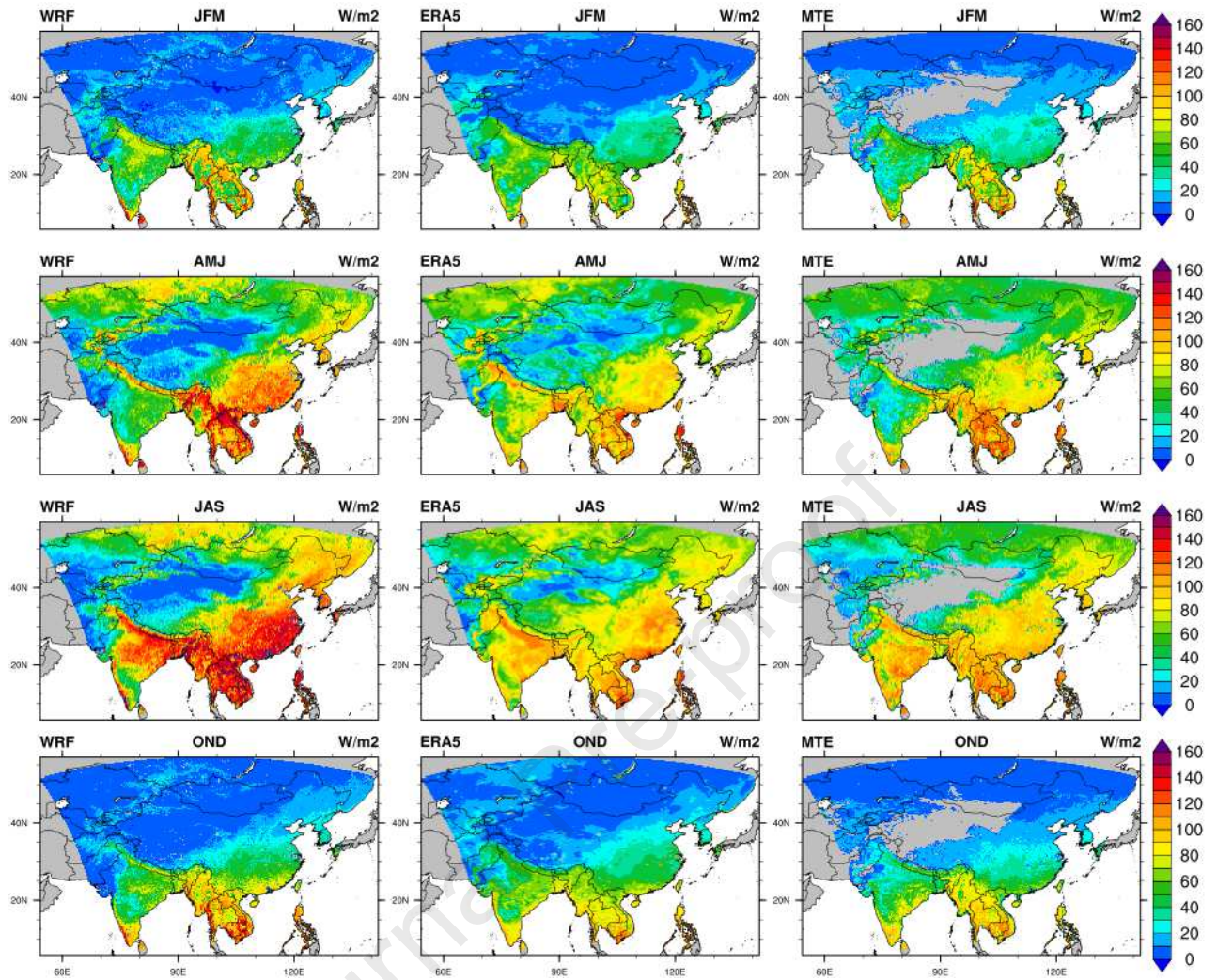
1247 **Figure 2.** Spatial pattern of precipitation (mm/day) as simulated by WRF-Chem (left panels)
 1248 and compared to ERA5 reanalysis (central panels) and CRU dataset (right panels) during
 1249 different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-August-
 1250 September, OND: October-November-December). Mind the differences in color scales
 1251 between seasonal climatologies.

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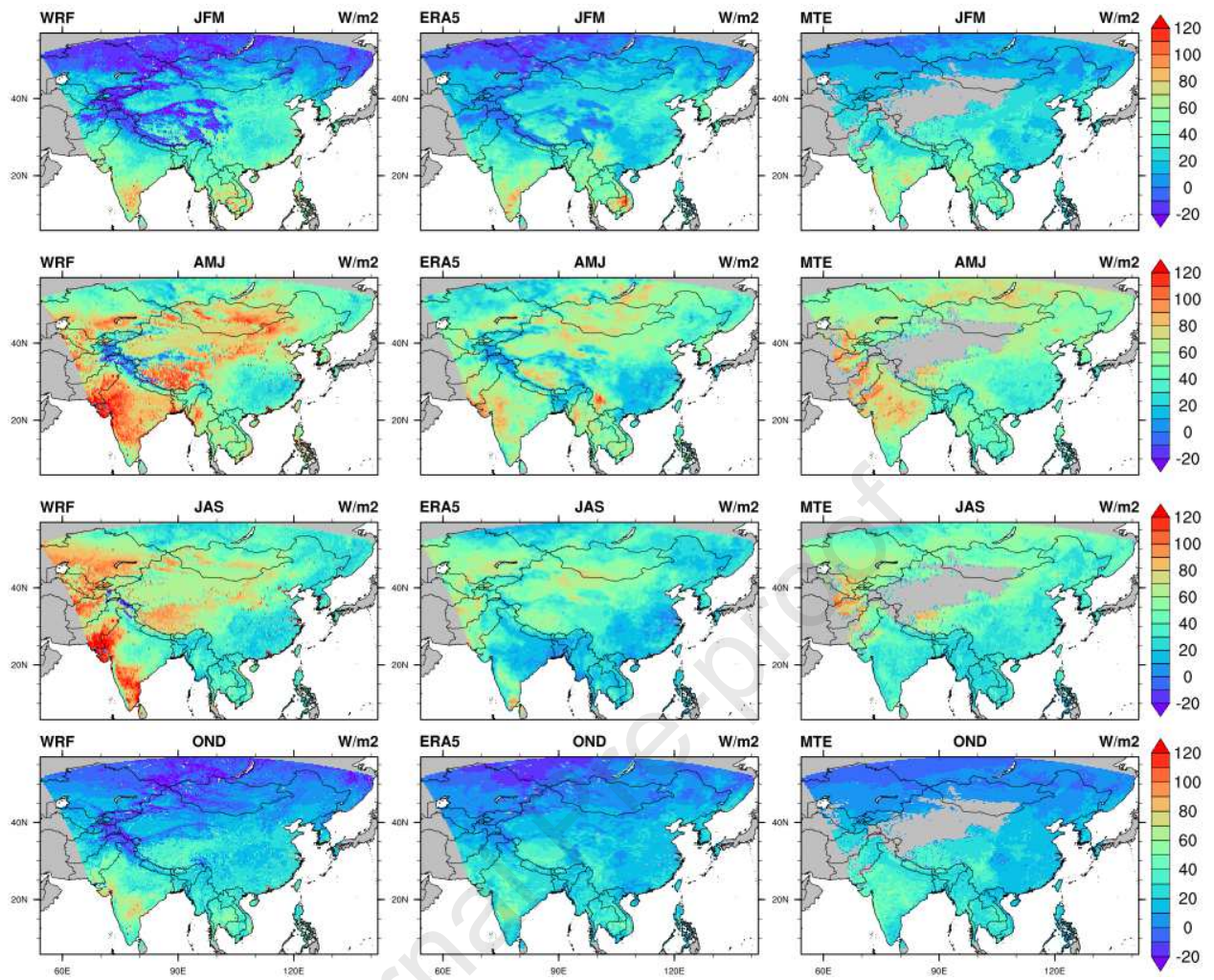
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1254 **Figure 3.** Spatial pattern of 2-meter relative humidity (%) as simulated by WRF-Chem (left
 1255 panels) and compared to ERA5 reanalysis (central panels) and CRU dataset (right panels)
 1256 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-
 1257 August-September, OND: October-November-December). Mind the differences in color
 1258 scales between seasonal climatologies.



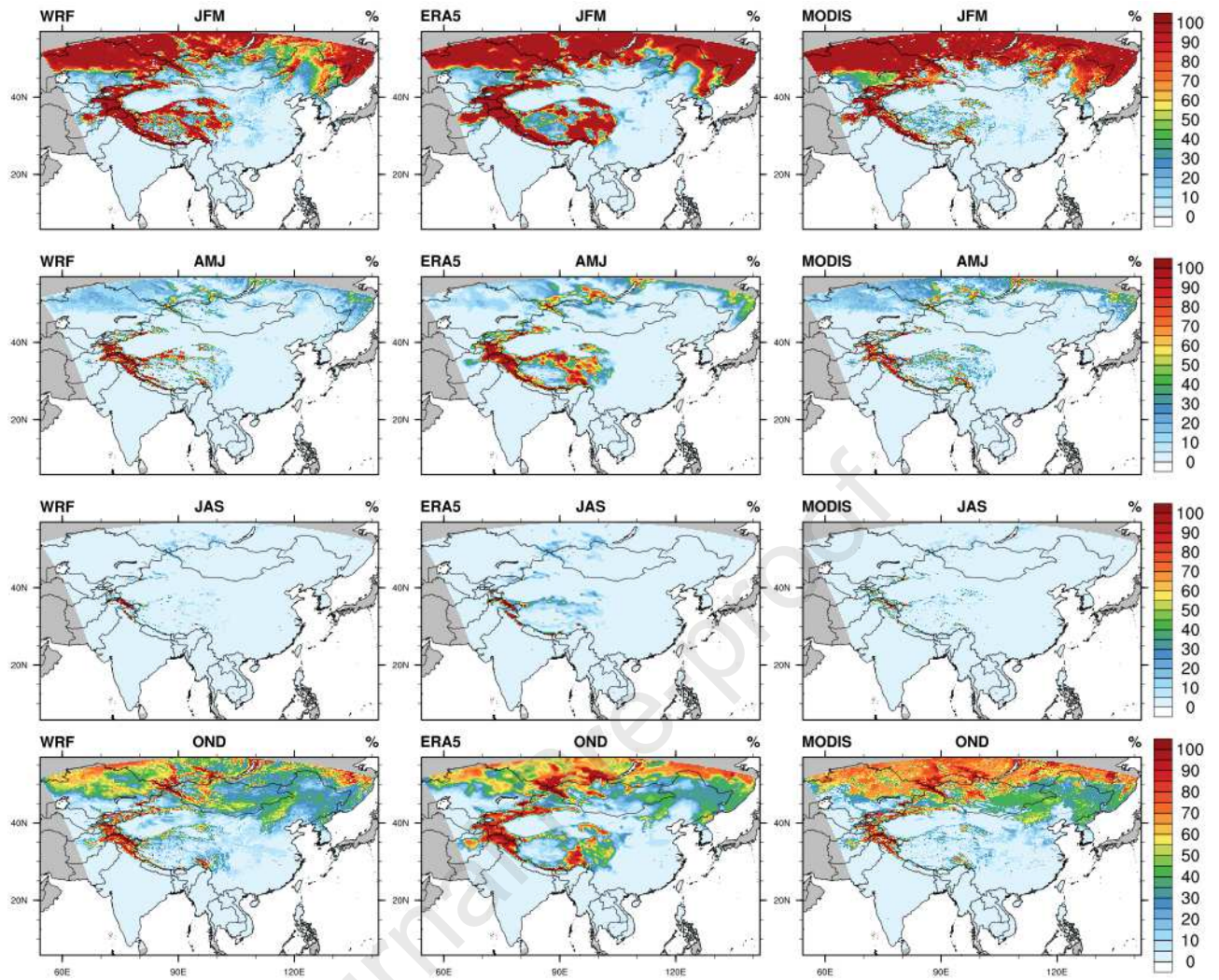
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1260 **Figure 4.** Spatial pattern of surface latent heat (W/m^2) as simulated by WRF-Chem (left
 1261 panels) and compared to ERA5 reanalysis (central panels) and CRU dataset (right panels)
 1262 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-
 1263 August-September, OND: October-November-December).



1264

1265 **Figure 5.** Spatial pattern of surface sensible heat (W/m²) as simulated by WRF-Chem (left
 1266 panels) and compared to ERA5 reanalysis (central panels) and CRU dataset (right panels)
 1267 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-
 1268 August-September, OND: October-November-December).

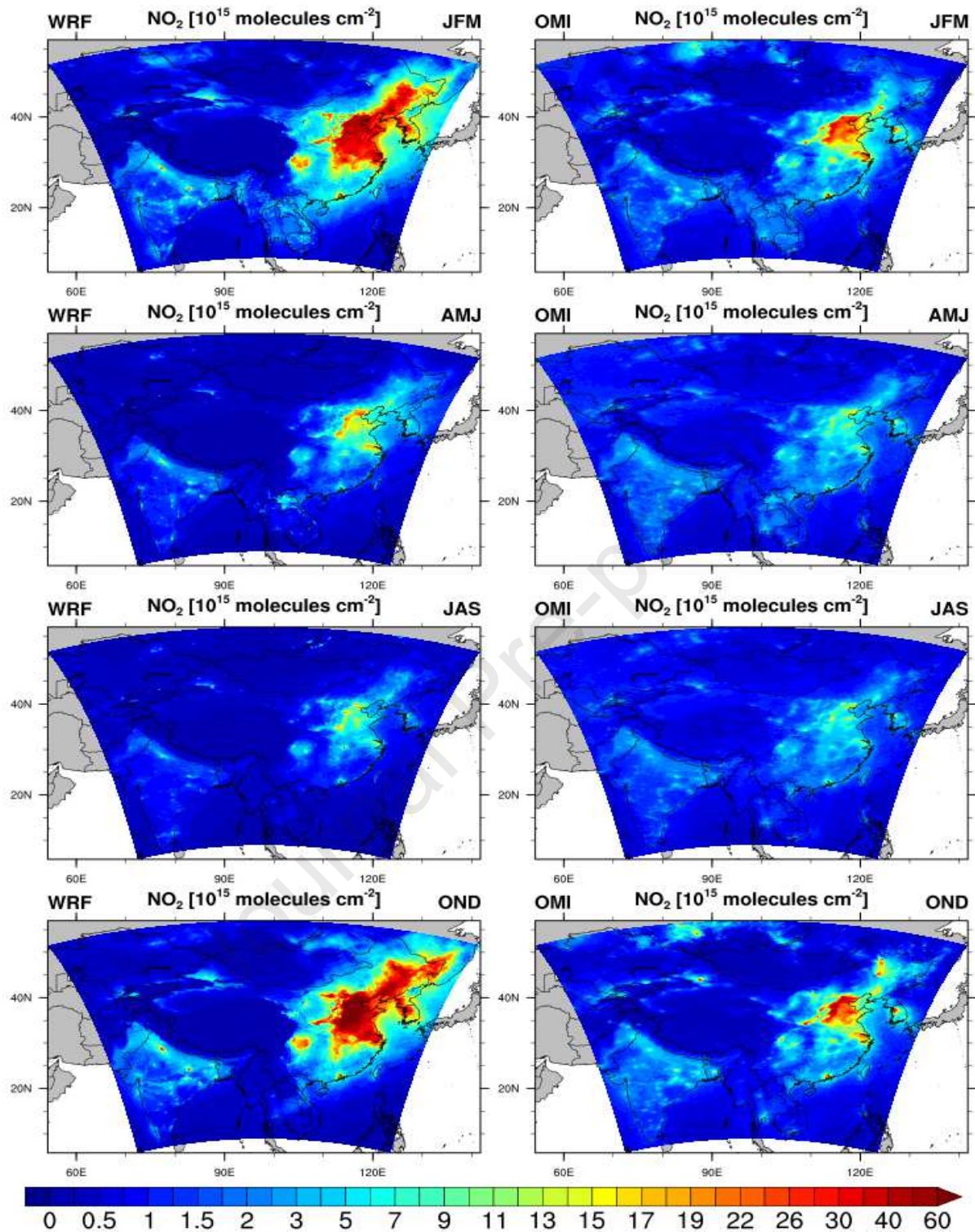


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1270 **Figure 6.** Seasonal spatial pattern of snow fractional cover (%) as simulated by WRF-Chem
 1271 (left panels) and compared to ERA5 (center panels) and MODIS data (right panels) during
 1272 different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-August-
 1273 September, OND: October-November-December).

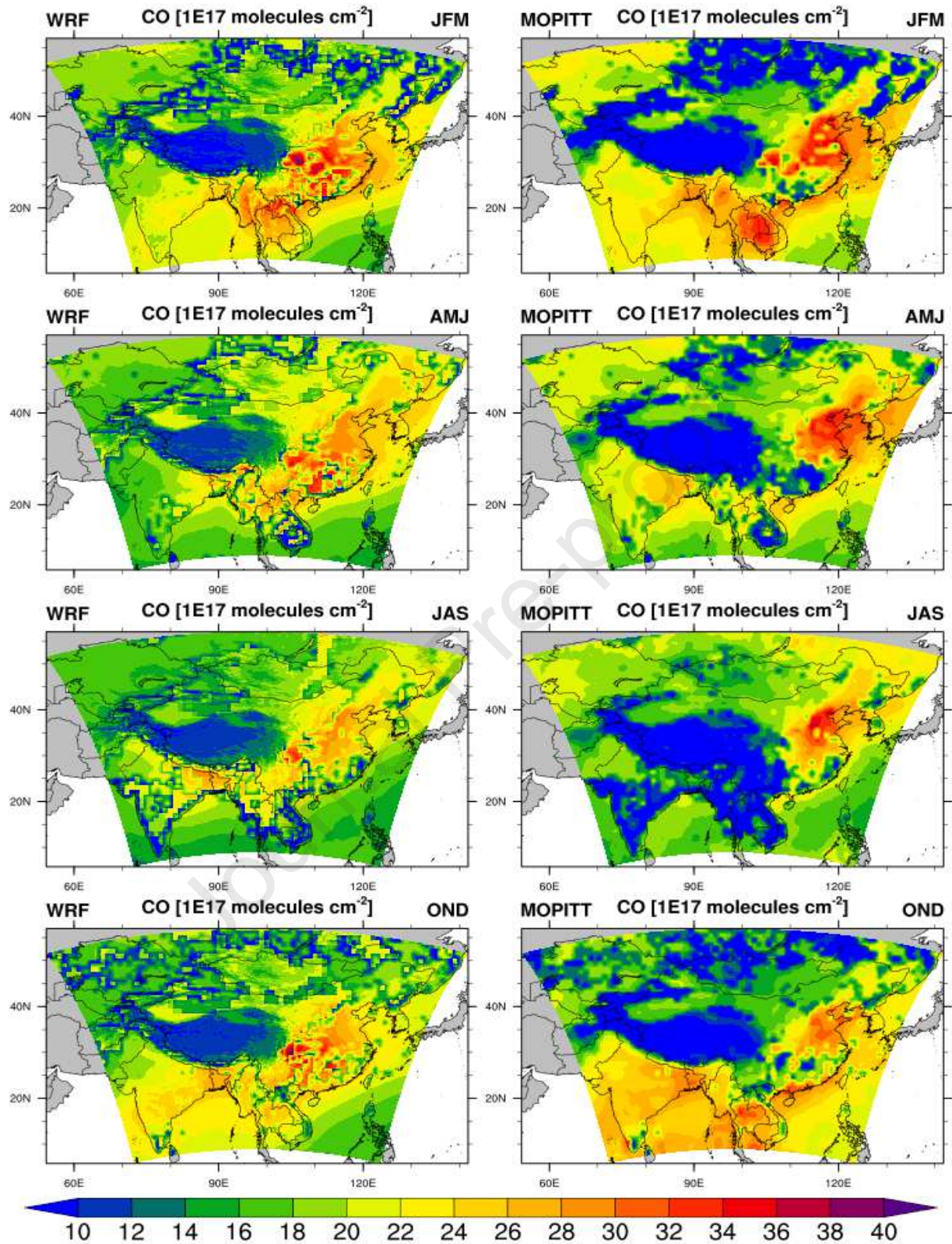
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1277 **Figure 7.** Spatial distributions of WRF-Chem simulated and OMI- retrieved tropospheric NO₂
 1278 content ($\times 10^{15}$ molecules/cm²) during different seasons (JFM: January-February-March,
 1279 AMJ: April-May-June, JAS: July-August-September, OND: October-November-December).

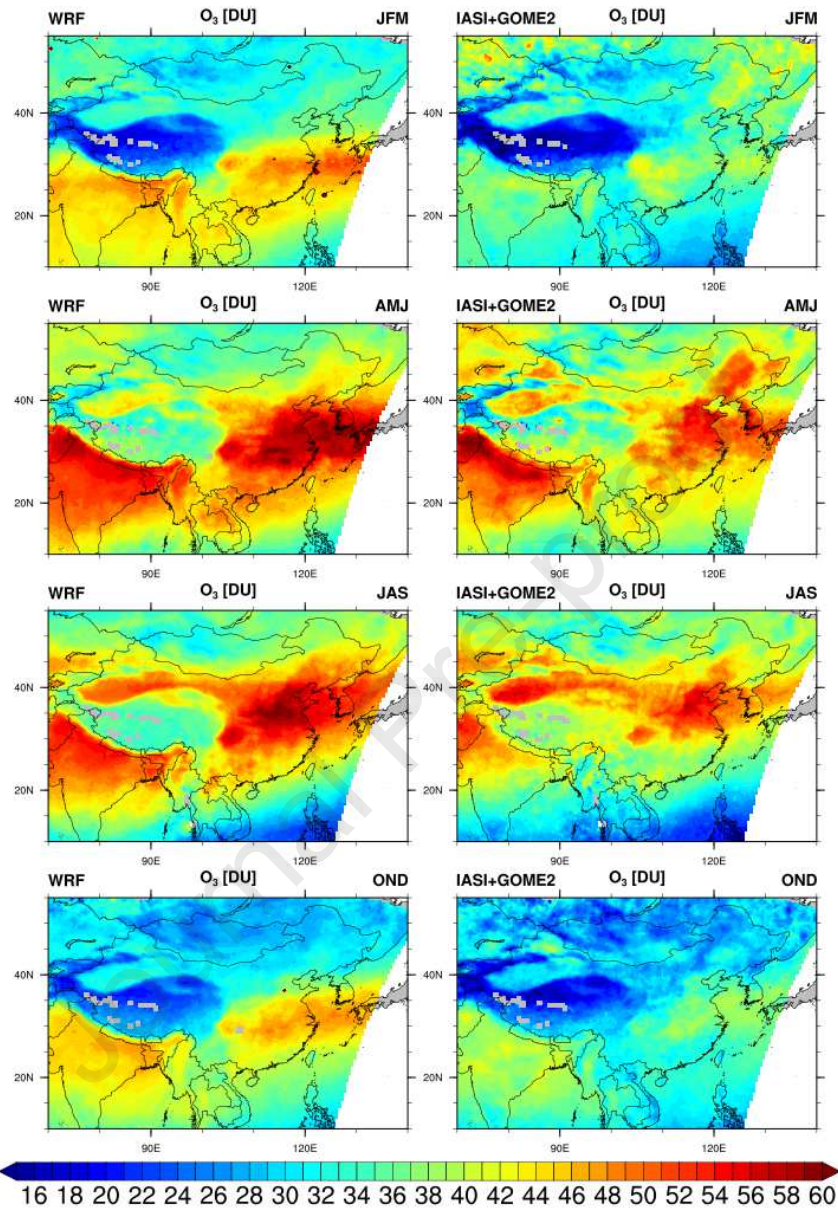


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1281 **Figure 8.** Comparison of WRF-Chem simulated total CO column with MOPITT data ($\times 10^{17}$
 1282 molecules/ cm^2) during different seasons (JFM: January-February-March, AMJ: April-May-
 1283 June, JAS: July-August-September, OND: October-November-December). The WRF-Chem
 1284 simulated CO are quite discontinuous due to coarse spatial resolution of MOPITT data (1x1
 1285 deg) to compute WRF total column.

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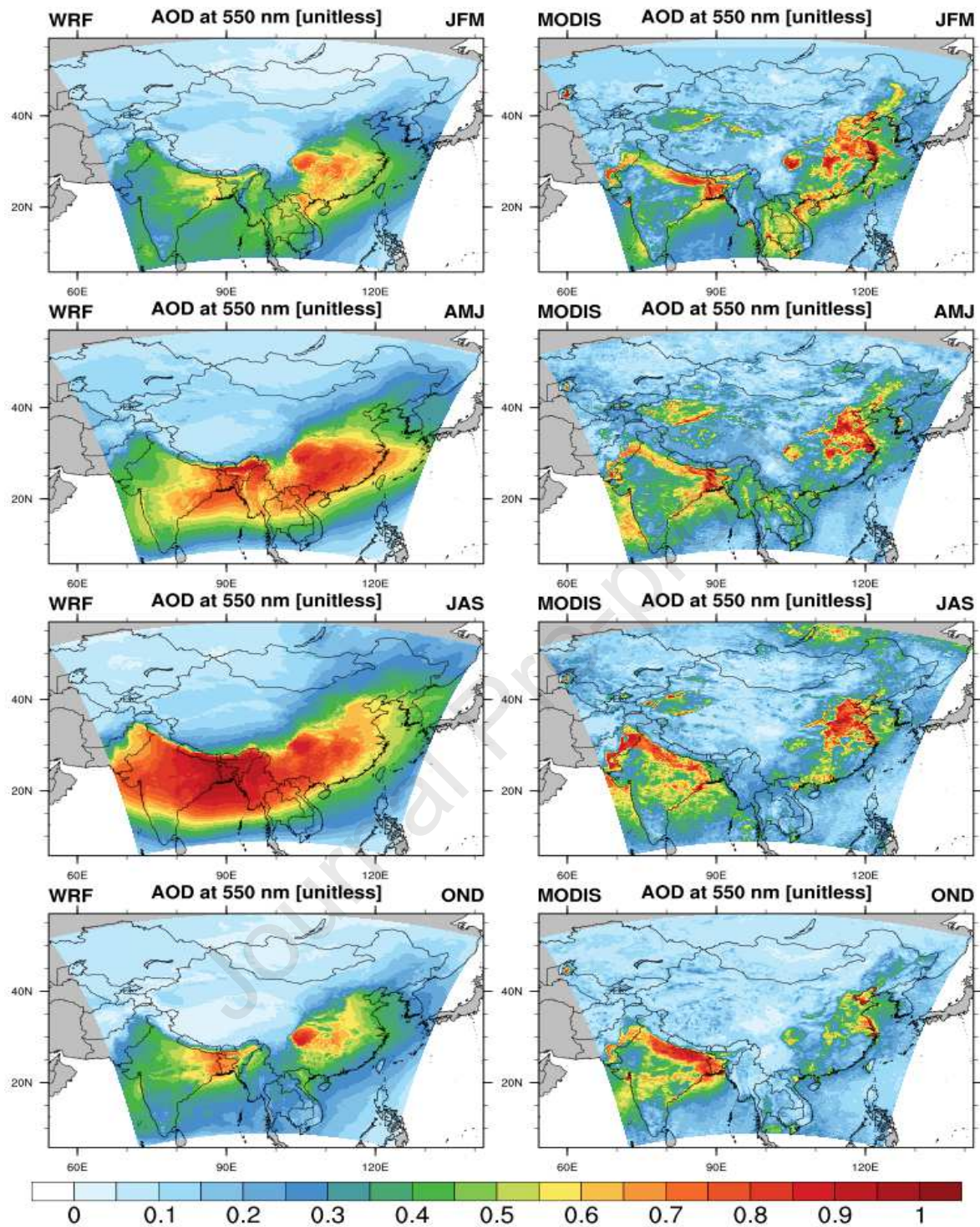
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1289 **Figure 9.** Spatial tropospheric column ozone (Dobson Unit, DU), integrated between the
 1290 surface and the tropopause, according to the WRF-Chem model (left panels) and IASI-
 1291 GOME2 satellite retrievals (right panels) during different seasons (JFM: January-February-
 1292 March, AMJ: April-May-June, JAS: July-August-September, OND: October-November-
 1293 December).

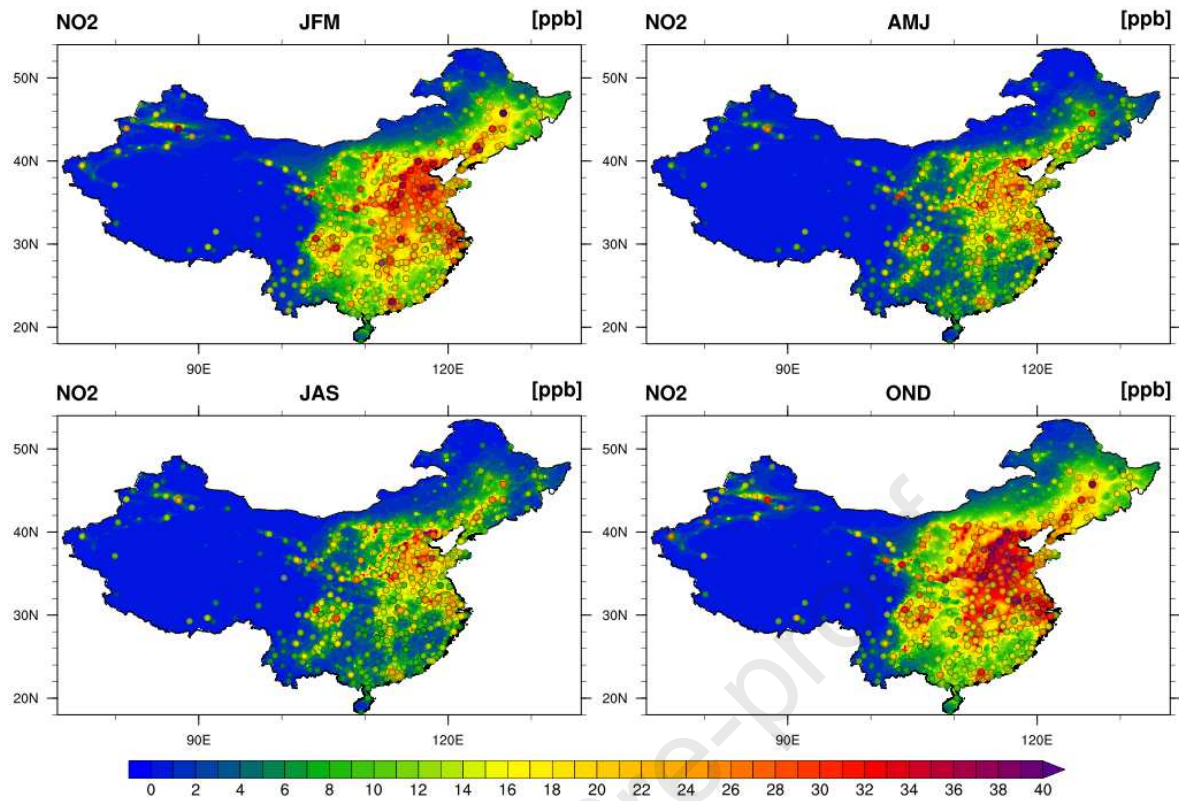
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1296 **Figure 10.** Comparison of simulated aerosol optical depth (dimensionless) with MODIS data
 1297 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS: July-
 1298 August-September, OND: October-November-December).

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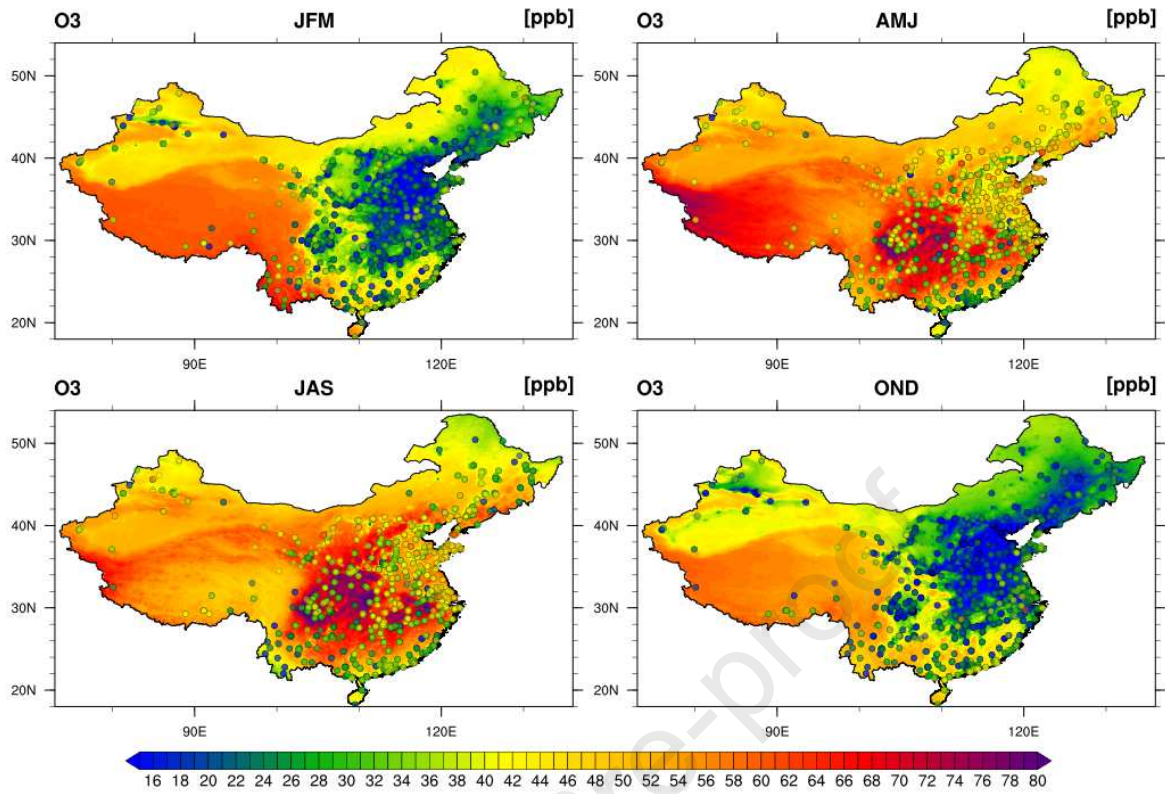


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1301 **Figure 11.** Spatial distributions of surface NO₂ concentrations (in ppb) simulated by the
1302 WRF-Chem model (background) and from air quality monitoring stations (dots) across China
1303 in 2015 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS:
1304 July-August-September, OND: October-November-December).

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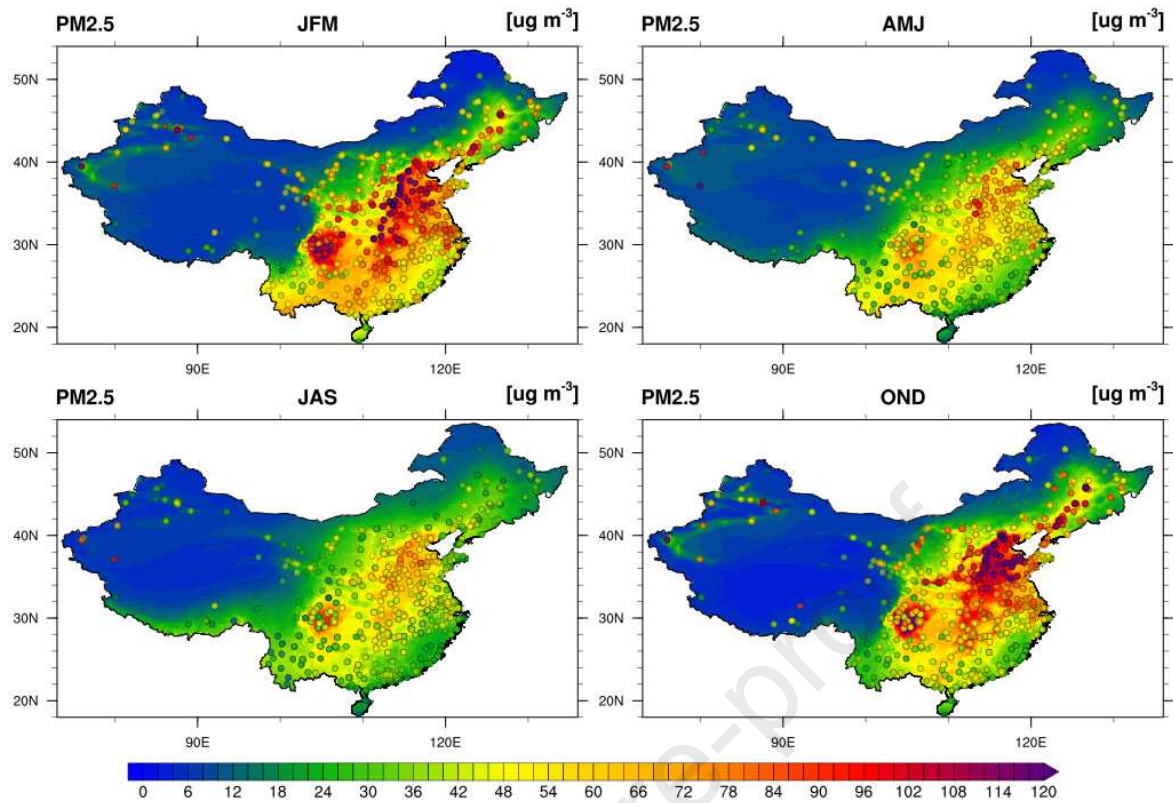
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1308 **Figure 12.** Spatial distributions of surface O₃ concentrations (in ppb) simulated by the WRF-
1309 Chem model (background) and from air quality monitoring stations (dots) across China in
1310 2015 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS:
1311 July-August-September, OND: October-November-December).

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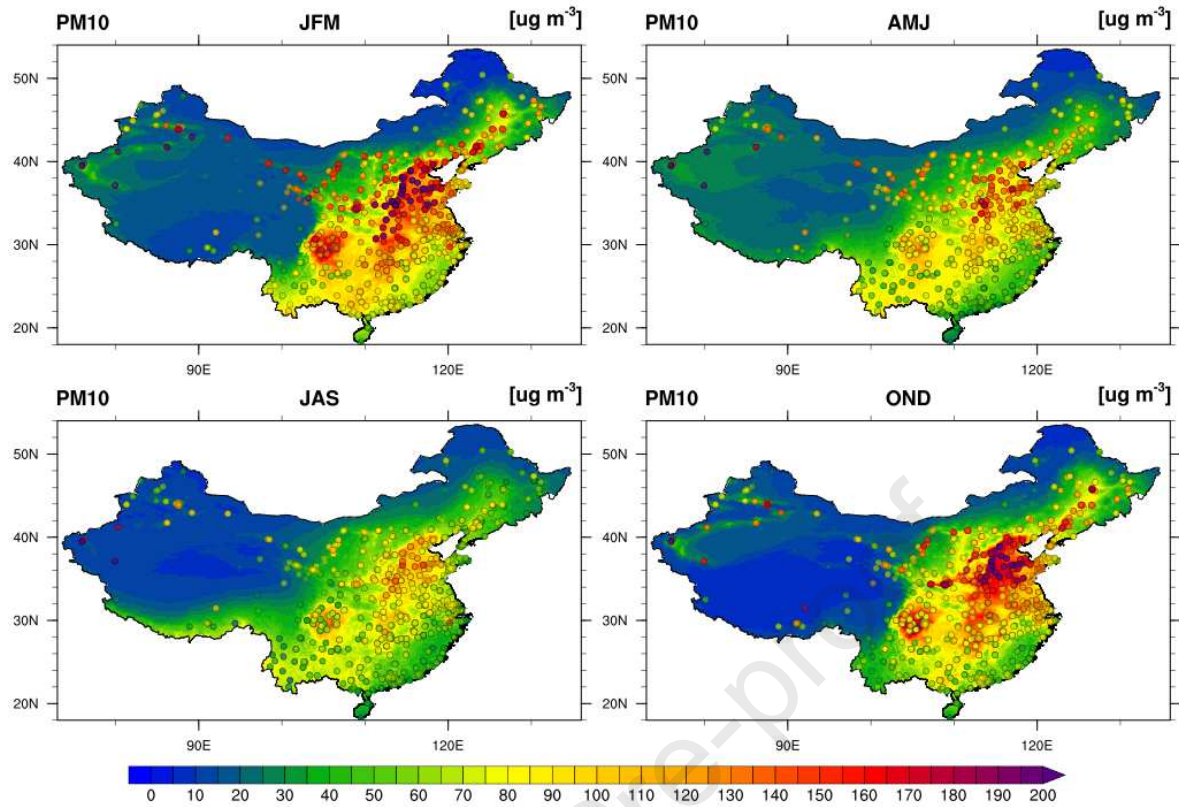


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1316 **Figure 13.** Spatial distributions of surface $\text{PM}_{2.5}$ concentrations (in $\mu\text{g m}^{-3}$) simulated by the
1317 WRF-Chem model (background) and from air quality monitoring stations (dots) across China
1318 in 2015 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS:
1319 July-August-September, OND: October-November-December).

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1323 **Figure 14.** Spatial distributions of surface PM₁₀ concentrations (in $\mu\text{g m}^{-3}$) simulated by the
1324 WRF-Chem model (background) and from air quality monitoring stations (dots) across China
1325 in 2015 during different seasons (JFM: January-February-March, AMJ: April-May-June, JAS:
1326 July-August-September, OND: October-November-December).

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Highlights

The WRF-Chem model was applied over Asia in 2015 at 8-km horizontal resolution

The outputs were evaluated against satellite and ground-based observations in China in 2015

WRF-Chem reproduced well the spatio-temporal patterns for meteorological and chemical variables

WRF-Chem reliable tool for air pollution risk assessment to human and ecosystems health

Journal Pre-proof

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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