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Research Paper

Evaluation and comparison of CMIP6 models and MERRA-2 reanalysis AOD against Satellite observations from 2000 to 2014 over China

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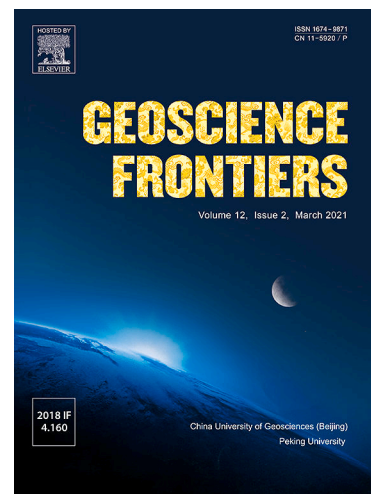
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1 **Research Paper**2 **Evaluation and comparison of CMIP6 models and MERRA-2 reanalysis**3 **AOD against Satellite observations from 2000 to 2014 over China**

4

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35

36 **Abstract**

37 Rapid industrialization and urbanization along with a growing population are contributing
38 significantly to air pollution in China. Evaluation of long-term aerosol optical depth (AOD)
39 data from models and reanalysis, can greatly promote understanding of spatiotemporal
40 variations in air pollution in China. To do this, AOD (550 nm) values from 2000 to 2014
41 were obtained from the Coupled Model Inter-comparison Project (CMIP6), the second
42 version of Modern-Era Retrospective analysis for Research, and Applications (MERRA-2),
43 and the Moderate Resolution Imaging Spectroradiometer (MODIS; flying on the Terra
44 satellite) combined Dark Target and Deep Blue (DTB) aerosol product. We used the Terra-
45 MODIS DTB AOD (hereafter MODIS DTB AOD) as a standard to evaluate CMIP6
46 Ensemble AOD (hereafter CMIP6 AOD) and MERRA-2 reanalysis AOD (hereafter
47 MERRA-2 AOD). Results show better correlations and smaller errors between MERRA-2
48 and MODIS DTB AOD, than between CMIP6 and MODIS DTB AOD, in most regions of
49 China, at both annual and seasonal scales. However, significant under- and over-
50 estimations in the MERRA-2 and CMIP6 AOD were also observed relative to MODIS
51 DTB AOD. The long-term (2000–2014) MODIS DTB AOD distributions show the highest
52 AOD over the North China Plain (0.71) followed by Central China (0.69), Yangtse River
53 Delta (0.67), Sichuan Basin (0.64), and Pearl River Delta (0.54) regions. The lowest AOD
54 values were recorded over the Tibetan Plateau (0.13 ± 0.01) followed by Qinghai ($0.19 \pm$
55 0.03) and the Gobi Desert (0.21 ± 0.03). Large amounts of sand and dust particles emitted
56 from natural sources (the Taklamakan and Gobi Deserts) may result in higher AOD in
57 spring compared to summer, autumn, and winter. Trends were also calculated for
58 2000–2005, for 2006–2010 (when China introduced strict air pollution control policies

59 during the 11th Five Year Plan or FYP), and for 2011–2014 (during the 12th FYP). An
60 increasing trend in MODIS DTB AOD was observed throughout the country during
61 2000–2014. The uncontrolled industrialization, urbanization, and rapid economic
62 development that mostly occurred from 2000 to 2005 probably contributed to the overall
63 increase in AOD. Finally, China's air pollution control policies helped to reduce AOD in
64 most regions of the country; this was more evident during the 12th FYP period (2011–2014)
65 than during the 11th FYP period (2006–2010). Therefore this study strongly advises the
66 authority to retain or extend these policies in the future for improving air quality.

67 **Keywords:** AOD, CMIP6, MERRA-2, MODIS, Trends

68

69 1. Introduction

70 Aerosols are mixtures of tiny solid and liquid suspended particles in the atmosphere,
71 having a standard radius from 0.001 to 100 μm . Aerosols are of great interest to the
72 scientific community due to their impact on global climate, radiative balance, air quality,
73 human health, cloud microphysical properties, hydrological cycle, ecosystems, and
74 agriculture (Charlson et al., 1992; Xia et al., 2006; Yu et al., 2009; Wang et al., 2015; You
75 et al., 2015; Shao et al., 2017). Several studies on this topic have been conducted over
76 different regions around the world to understand aerosol behavior (Che et al., 2009, 2015a,
77 2015b; Zhang et al., 2012; Bilal and Nichol, 2015, 2017; Kong et al., 2016; Ma et al., 2016;
78 Xin et al., 2016; Bilal et al., 2017a, 2017b, 2018; Ali et al., 2019, 2020; Zhang et al., 2020;
79 Su et al., 2021). Even so, aerosol optical properties remain an important source of
80 uncertainty in climate projection and assessment (IPCC, 2013). Aerosol optical depth
81 (AOD) is an essential parameter that measures aerosol loading in the atmosphere and is a

82 principal factor for climate change research (Pan et al., 2010). Therefore, AOD evaluation
83 at local to global scales, its long-term spatiotemporal variations, and trend calculations, are
84 necessary, and especially in China which has severe air pollution.

85 Such analysis requires multiple datasets (e.g., ground measurements, satellite remote
86 sensing, reanalysis, and model simulation) for a comprehensive understanding of the nature
87 and effects of aerosols. National Aeronautics and Space Administration (NASA) Aerosol
88 Robotic Network (AERONET) (Holben et al., 1998), in 2001. Complementary to
89 AERONET, and in part shared with AERONET, Chinese networks have been developed
90 (Xia et al., 2021), such as CARSNET (China Aerosol Remote Sensing Network),
91 established by the China Meteorological Administration (CMA) (Che et al., 2009, 2015a)
92 (50 sites), and SONET (Sun-Sky Radiometer Observation Network) established by the
93 Institute of Remote Sensing and Digital Earth, Chinese Academy of Sciences (RADI/CAS),
94 Beijing (Li et al., 2018) (16 sites). CARE China (Campaign on Atmospheric Aerosol
95 Research network of China) (Xin et al., 2015) includes 36 sites where handheld sun
96 photometers are used. The limitation of these networks is their sparse spatial distribution
97 (Holben et al., 2001). This limitation can be overcome by satellite remote sensing
98 techniques and Global Climate Models (GCMs), which provide wide spatial coverage of
99 AOD at local to global scales. AOD retrievals are available from several satellite-based
100 instruments such as the Total Ozone Monitoring Instrument (TOMS) (Torres et al., 2002),
101 Advanced Very High-Resolution Radiometer (AVHRR) (Hauser, 2005), the Along Track
102 Scanning Radiometers (ATSR-2 and AATSR) (Holzer-Popp et al., 2013; de Leeuw et al.,
103 2015; Popp et al., 2016), Ozone Monitoring Instrument (OMI) (Torres et al., 2007), Multi-
104 angle Imaging Spectroradiometer (MISR) (Kahn et al., 2010), the Sea-Viewing Wide-field
105 of View Sensor (SeaWiFS) (Sayer et al., 2012), the Visible Infrared Imaging Radiometer
106 Suite (VIIRS) (Liu et al., 2014), and the Moderate Resolution Imaging Spectroradiometer

107 (MODIS) (Remer et al., 2005; Hsu et al., 2006, 2013; Levy et al., 2010, 2013; Sayer et al.,
108 2014). In addition, reanalysis data such as from Copernicus Atmosphere Monitoring
109 Services (CAMS) (Flemming et al., 2017), the second Modern-Era Retrospective analysis
110 for Research and Applications (MERRA-2) (Randles et al., 2017), and Global Climate
111 Models (GCMs) such as the Coupled Model Intercomparison Project Phase6 (CMIP6)
112 (Eyring et al., 2016) also provide long-term spatial AOD data.

113 MODIS, onboard the Terra and Aqua satellites is one of the most widely appraised
114 instruments in retrieving AOD (Levy et al., 2013; Sayer et al., 2014; Bilal and Nichol,
115 2015; Mhawish et al., 2017). The satellite-based MODIS sensor provides global AOD
116 distributions using the dark target (DT) and deep blue (DB) algorithms, which have been
117 widely evaluated over vegetated and bright reflecting surfaces (Kaufman et al., 1997; Hsu
118 et al., 2013). In addition, several researchers evaluated the MODIS DT, DB, and their
119 combined DTB AOD products over different regions (Chu et al., 2002; Bilal et al., 2016,
120 2017b, 2018, 2021a; Georgoulas et al., 2016; Nichol and Bilal, 2016; Ali et al., 2017; Butt
121 et al., 2017; Mhawish et al., 2017; Wang et al., 2017; de Leeuw et al., 2018; Sogacheva et
122 al., 2018, 2020; Ali and Assiri, 2019; Almazroui, 2019; Bright and Gueymard, 2019; Mei et
123 al., 2019; Tian and Gao, 2019; Filonchik and Hurynovich, 2020; Wei et al., 2020). Apart
124 from these, several studies also evaluated MODIS AOD products against ground
125 measurements in different regions of China. For example, Wang et al. (2019) evaluated
126 MODIS collection (C6.1) DT and DB AOD against 20 AERONET sites over different
127 regions in China and reported that DB performed well to estimate AOD in terms of
128 correlation coefficient ($r = 0.931$), root mean squared error ($RMSE = 0.18$), relative mean
129 bias ($RMB = 1.02$), and an acceptable percentage of retrievals within the expected error
130 ($EE = 63.49\%$). The corresponding DT values were $r = 0.946$, $RMSE = 0.19$, $RMB = 1.17$,
131 and $EE = 54.03\%$. The MODIS C6.1 DB AOD performed better against AERONET AOD

132 measurements over Beijing, XiangHe, and Xinglong sites as indicated by r (0.92), RMSE
133 (0.22), and EE (68.3%) compared to the DT algorithm (Bilal et al., 2019). Huang et al.
134 (2020) evaluated Terra and Aqua based MODIS C6.1 DT and DB AOD against SONET
135 sites in the westernmost city (Kashi region, Xinjiang Uygur Autonomous Region) in China
136 and reported that DB showed better results for both satellites in terms of r (Terra/Aqua =
137 0.896/0.907), RMSE (0.283/0.203), RMB (0.337/0.388), and EE (82.35/84.06%) compared
138 to the DT algorithm. In addition, Che et al. (2019a) validated MODIS C6 and C6.1 DT
139 AOD over 18 AERONET and CARSNET sites in China for the period 2002 to 2014. They
140 found that DT C6.1 obtained better results in retrieving AOD in terms of r (0.901), RMSE
141 (0.171), RMB (0.998), and EE (59.03%) compared to the C6 DT algorithm ($r = 0.890$,
142 RMSE = 0.185, RMB = 1.039, EE = 54.94%). Huang et al. (2019) validated MODIS C006
143 and 61 DT AOD 3km products against AERONET sites in China, and they reported that
144 C6.1 DT performed better in terms of R^2 (0.87), RMSE (0.23), RMB (1.41), and EE (45%)
145 than did C6 DT (0.81, 0.31, 1.57, and 39%). Apart from these, Li et al. (2020) reported that
146 the C6.1 MODIS DTB AOD product produced better results over 12 AERONET sites in
147 China as indicated by R^2 (Terra/Aqua = 0.81/0.79), RMSE (0.15/0.17), and EE
148 (66.63/65.32%) compared to the DT and DB algorithms. The DTB AOD product also has
149 good spatial coverage over multilayer surfaces in China. Filonchik et al. (2019) used this
150 product for a local study and found that the combined C6.1 MODIS DTB AOD performed
151 better over four AERONET sites (Beijing, XiangHe, Taihu, and SACOL), as indicated by r
152 (0.885–0.902), RMSE (0.097–0.302), RMB (0.97–1.17), and EE (76.3%–78.3%). Based
153 on the good performance of the DTB AOD retrievals over China, as reported by previous
154 studies, the present study used the combined Terra-MODIS C6.1 DTB AOD as the
155 reference data to evaluate CMIP6 and the MERRA-2 AOD.

156 Very few studies were found that evaluated CMIP5/CMIP6 models and MERRA-2
157 reanalysis-based AOD against satellite observations from local to global scales. Mortier et
158 al. (2020) calculated AOD trends using the CMIP6 and AeroCom models at a global scale
159 and reported decreasing trends in AOD over Europe, North and South America, and North
160 Africa. Li et al. (2020) evaluated the CMIP5 model-based AOD against MODIS AOD over
161 East Asia from 2001 to 2005 and reported an underestimation in CMIP5 AOD. Similar
162 studies were conducted by Misra et al. (2016) and Sockol and Small Griswold (2017) over
163 the USA and India, respectively. In China, Sun et al. (2019) evaluated the MERRA-2
164 reanalysis AOD against Aqua-based MODIS AOD from 1980 to 2010 and Liu et al. (2021)
165 evaluated CAMS and MERRA-2 AOD against MODIS AOD over the Sichuan Basin of
166 China. However, no comprehensive evaluation of MERRA-2 and CMIP6 AOD data was
167 found over China. Due to the unprecedented social and economic developments of China in
168 recent decades, the country now suffers from intense aerosol pollution (Wang et al., 2021).
169 In 2015, a total of 1710 days of severe and above pollution, and 154 heavy pollution
170 weather warnings were observed across 70 cities in the BTH region. In 2016, Yale
171 University published the Environmental Performance Index (EPI) report, which ranked
172 China as having the second worst air quality globally, before Bangladesh (Song et al.,
173 2019; Qiu et al., 2021). For the use of MERRA-2 reanalyses and CMIP6 AOD for air
174 quality assessments, a comprehensive evaluation of the model results is required to see
175 how well they represent the local to regional spatio-temporal AOD scenarios and trends
176 over China. In this contribution, the model results are evaluated against MODIS AOD. The
177 main objectives of this study are: (1) to evaluate CMIP6 Ensemble AOD and MERRA-2
178 reanalysis AOD against Terra-MODIS AOD; (2) investigate the long-term spatiotemporal
179 discrepancy of AOD at annual and seasonal scales; and (3) to estimate the effect of
180 China's air pollution control policies on AOD.

181

182 **2. Data and methods**183 *2.1 Study area*

184 In this study, AOD from multiple sources was investigated over China. China is
185 characterized by having a diverse climate and broad geography, and is located at 3°51'–53°
186 33' N latitudes and 73°33'–135° 05' E longitudes (Fig. 1). China's regions have a large
187 landmass with a complex topography and aerosol heterogeneity. Climatologically, China's
188 climate varies from south and southeast (humid) to north and northwest (dry), with an
189 uneven pattern of precipitation, which is mainly due to the distance from the sea. The
190 northern and western regions of the country are dominated by the Gobi and Taklamakan
191 Deserts with plateaus and massifs, whereas the southern regions comprise hilly and
192 mountainous terrain. The southern coastal and eastern plains are composed of fertile
193 lowlands and foothills, and so the largest number of people live in these regions. The
194 country has four distinct seasons. December to February (DJF) represent a harsh cold
195 winter with a very dry climate in most of China, except for southern China, which remains
196 just cool. A more moderate climate occurs in spring from March to May (MAM), while the
197 summer months June to August (JJA) brings are very hot and humid. The autumn lasts
198 from September to November (SON) and is characterized by warm weather with strong
199 winds and infrequently heavy rain.

200 In China, aerosol distribution varies over time and from region to region; therefore a
201 region-based study is more effective in distinguishing the long-term changes in aerosols
202 compared to the national scale (Zhao et al., 2008). Emissions from different industries
203 located in different regions are key sources of aerosol in China. Industrial aerosols are
204 emitted mostly from the Eastern (Pan et al., 2010; Deng et al., 2012; He et al., 2016; Kang

205 et al., 2016a, 2016b) and Central (Wang et al., 2015; Liu et al., 2016) part of China. In
206 addition, dust aerosols across the Northern and Northwestern regions of China are triggered
207 by wind erosion (Tan et al., 2015; Yu et al., 2016). Marine aerosols found over the Eastern
208 and Southern parts of China are generated from the Yellow Sea and the South China Sea
209 (Kang et al., 2016a). In light of the above, ten major regions (see Fig.1) were selected
210 across China. Results for the ten regions are amalgamated to calculate results for an 11th
211 region “Entire China”, representing the whole of China.

212

213 *2.2 Aerosol products*

214 *2.2.1 CMIP6 based AOD datasets*

215 In this study, we used multi-model historical simulations from the latest CMIP6
216 archive, which extend up to 2014. Trends in AOD at 550 nm for the 10 selected regions in
217 China were investigated for the 2000–2014 simulation period. In this regard, we used AOD
218 data from 15 CMIP6 up-to-date global climate models (GCMs). Based on the availability
219 of required data, output from those 15 models was downloaded from the CMIP6 website.
220 Some details of the selected 15 GCMs are summarized in Table 1.

221

222 *2.2.2 MERRA-2 reanalysis AOD datasets*

223 In this study, the reanalysis AOD from MERRA-2 at a spatial resolution of $0.5^\circ \times$
224 0.625° was used. MERRA-2 is the updated version of the original MERRA reanalysis
225 datasets (Rienecker et al., 2011). The inclusion of the Goddard Earth Observing System
226 (GEOS) model, as well as the assimilation of observation types, is the fundamental
227 enhancement in this upgraded version (Molod et al., 2015; Georgoulias et al., 2016; Gelaro

228 et al., 2017). MERRA-2 is recognized as the first satellite reanalysis product to assimilate
229 aerosol information for the earth system. It represents the interaction between aerosols and
230 climate system variables on the Earth. The aerosol model is developed based on the
231 assimilation of AOD products from the Advanced Very High-Resolution Radiometer
232 (AVHRR), MISR (Multi-angle Imaging Spectroradiometer), and MODIS satellite-based
233 sensors, as well as from ground-based observations such as provided by AERONET
234 (Aerosol Robotic Network). For more details about MERRA-2 and the evaluation of
235 aerosol assimilation, see Gelaro et al. (2017), Randles et al. (2017), and Shi et al. (2019).

236 2.2.3 MODIS AOD datasets

237 This study also used the MODIS aerosol products, as they have the greater availability
238 of effective AOD pixels (de Meij et al., 2012; de Leeuw et al., 2018). To explore the
239 spatiotemporal features of aerosols, MODIS was launched in 1999 onboard the Terra
240 (descending orbit, local crossing time: 10:30 AM) satellite and in 2002 onboard the Aqua
241 (ascending orbit, local crossing time: 01:30 PM) satellite as part of NASA's Earth
242 Observing System (EOS) mission. This sensor measures the upwelling Earth radiation in 36
243 spectral channels from 0.4–14.4 μm and with three different spatial resolutions (e.g., 250 m
244 for bands 1–2, 500 m for bands 3–7, 1 km for 8–36) with a swath viewing of 2330 km (for
245 details see <https://modis.gsfc.nasa.gov/about/specifications.php>; accessed date: 22 Feb
246 2021). To retrieve MODIS AOD, three different algorithms, i.e. the dark target (DT) land
247 algorithm, the dark target (DT) ocean algorithm, and the deep blue (DB) land algorithms
248 are used (Hsu et al., 2013; Levy et al., 2013; Remer et al., 2013). The DT algorithm is
249 applicable for vegetated surfaces, whereas the DB algorithm is used for both bright
250 reflecting (desert surface) and vegetated surfaces. However, based on upgrades to the
251 algorithms, MODIS AOD datasets are stored at different levels and versions (known as

252 collections). The collection (C6.1) is the latest version of DT and DB algorithms, where
253 significant improvements and modifications were implemented from the previous C51 and
254 C6 versions (Levy et al., 2013; Bilal and Nichol, 2015; Bilal et al., 2016; Georgoulias et al.,
255 2016; Nichol and Bilal, 2016), as described in Sayer et al. (2019) and initially validated
256 over China by Sogacheva et al. (2018). This study used combined MODIS AOD products,
257 considering their reliability and extended coverage in terms of the enormous number of
258 valid pixels and their quality in retrieving AOD over both land and ocean surfaces (Levy et
259 al., 2013; Sayer et al., 2014). The MODIS C6.1 combined DT and DB (DTB) monthly
260 AOD (at 550 nm), from the Terra satellite with a spatial resolution of $1^\circ \times 1^\circ$, was obtained
261 from NASA Giovanni (<https://giovanni.gsfc.nasa.gov/giovanni/>; accessed date: 20
262 December 2020). Several studies were also used level 3 MODIS DTB AOD with a spatial
263 resolution of $1^\circ \times 1^\circ$ to investigate air pollution scenario (Ali and Assiri, 2016; Ali et al.,
264 2017, 2019; de Leeuw et al., 2018; Nichol et al., 2020; Qiu et al., 2021). More detailed
265 statistics about MODIS, its products, calibration process, retrieval algorithms, and
266 associated uncertainties have been discussed elsewhere (Levy et al., 2015; Sayer et al.,
267 2015; Georgoulias et al., 2016; Bilal et al., 2017b, 2018; Bilal and Nichol, 2017; de Leeuw
268 et al., 2018; Ali and Assiri, 2019). Henceforth, the Terra MODIS C6.1 combined DTB
269 product is referred to as MODIS DTB AOD.

270 *2.3 Research Methodology*

271 We followed these step-by-step methods to achieve our objectives:

- 272 • The AOD obtained from 15 CMIP6 models, MERRA-2 reanalysis, and Satellite (Terra
273 MODIS DTB) was interpolated onto the same geographical grid ($0.5^\circ \times 0.5^\circ$) using the
274 bilinear interpolation technique (Yousefi et al., 2020; Wang et al., 2021) in the Climate
275 Data Operators (CDO) tool.

- 276 • To reduce the differences among the 15 CMIP6 models and ensure the accuracy of
 277 AOD changes, a new AOD dataset (i.e., CMIP6 Ensemble) was generated from the
 278 combination of 15 CMIP6 models using the CDO tool. The study used a nearest
 279 interpolation technique using MATLAB software to remove the data gaps that affect
 280 the results and analysis of the MODIS AOD products (Yang and Hu, 2018).
- 281 • Several statistical methods were used to evaluate the CMIP6 and MERRA-2 AOD
 282 against MODIS DTB AOD. The methods are as follow:

283 The linear regression technique (Wilks, 2007) was used to calculate slope, intercept, and
 284 significance (Eq. (1)):

$$285 \quad Y = mx + c \quad (1)$$

286 where Y is the linear estimate, m defines the slope (a change of Y per unit changes of x),
 287 and c indicates the intercept or constant. The slope value is defined as the trend of the
 288 CMIP6, MERRA-2, and MODIS-DTB based AOD. Trend significance is estimated based
 289 on the p-value (a null hypothesis) (Ali et al., 2019). A p-value less than or equal to (≤ 0.05)
 290 is defined as significant with a 95% confidence level, while $p > 0.05$ indicates statistically
 291 insignificant. In addition, to calculate the uncertainty of CMIP6 and MERRA-2 AOD, we
 292 used the Pearson's correlation (r), root mean squared error (RMSE), mean absolute error
 293 (MAE), relative mean bias (RMB) (Bilal et al., 2016, 2021a, 2021b; Ali and Assiri, 2019;
 294 Almazroui, 2019; Islam et al., 2019)

$$r = \frac{\sum_{i=1}^n (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^n (x_i - \bar{x})^2 \sum_{i=1}^n (y_i - \bar{y})^2}} \quad (2)$$

$$\text{RMSE} = \sqrt{\frac{1}{n} \sum_{i=1}^n (\text{Model AOD} - \text{MODIS DTB AOD})^2} \quad (3)$$

$$\text{MAE} = \frac{1}{n} \sum_{i=1}^n |(\text{Model AOD} - \text{MODIS DTB AOD})| \quad (4)$$

$$\text{RMB} = \left(\frac{\text{Model AOD} - \text{MODIS DTB AOD}}{\text{MODIS DTB AOD}} \right) \quad (5)$$

295 where Model = CMIP6 and MERRA-2, RMB = 1 defines the normal estimation of CMIP6
 296 and MERRA-2, and positive and negative values represent over- and underestimations,
 297 respectively.

- 298 • Spatial and area-averaged maps of mean annual and seasonal AOD were generated from
 299 monthly CMIP6, MERRA-2, and MODIS DTB AOD from 2000–2014.

300

301

302 **3. Results and discussion**

303 *3.1 Evaluation of CMIP6 and MERRA-2 AOD against MODIS DTB AOD*

304 Figure 3 shows that the better performance of the MERRA-2 AOD with respect to the
 305 MODIS DTB AOD, than that of the CMIP6 Ensemble AOD. The MERRA-2 statistical
 306 measures showed higher correlation ($r = 0.71\text{--}0.89$) and lower error (RMSE = 0.07–0.23,
 307 MAE = 0.05–0.17) than that of the CMIP6 Ensemble AOD with a lower correlation ($r =$
 308 $0.38\text{--}0.66$) and higher error (RMSE = 0.05–0.28, MAE = 0.04–0.23) over all regions as
 309 well as for Entire China. In terms of RMB, both datasets underestimated AOD (MERRA-2
 310 = -0.05 to -0.22 , CMIP6 = -0.05 to -0.30) over the Northeast, North China Plain, and
 311 Pearl River Delta and overestimate AOD (MERRA-2 = 0.11, CMIP6 = 0.53) over the Gobi
 312 Desert except for the Tibetan Plateau. MERRA-2 performed better than CMIP6, with
 313 moderate correlations ($r = 0.51\text{--}0.64$) and errors (RMSE = 0.11–0.20, MAE = 0.08–0.15)
 314 over the Yangtse River Delta, Central, Qinghai, and Tarim Basin of China. The RMB
 315 values demonstrate that both datasets underestimated AOD over the Yangtse River Delta
 316 and the Central region (MERRA-2 by -0.08 to -0.19 , CMIP6 by -0.02 to -0.16) and

317 overestimated AOD (MERRA-2 by 0.01, CMIP6 by 0.60) over the Tarim Basin. However,
318 MERRA-2 underestimated (-0.36) and CMIP6 overestimated AOD (0.17) over Qinghai.
319 Over the Sichuan Basin, both datasets performed poorly due to significant overestimation
320 of lower AOD values, and underestimation of higher ones, which can explain the lower
321 correlations and higher errors.

322

323 Seasonally, the evaluation results show that MERRA-2 provides more accurate AOD
324 results at local scales than CMIP6 (Figs. 4 and 5). Specifically, over the Gobi Desert, for
325 MERRA-2 the correlation is higher ($r = 0.77$) and the error metrics are lower (RMSE =
326 0.06, MAE = 0.05) in the summer than in the winter, autumn, and spring, whereas for
327 CMIP6 the correlation is highest ($r = 0.30$) and the error metrics are smaller (RMSE = 0.11,
328 MAE = 0.10) in the summer than in the spring, winter, and autumn. CMIP6 tends to
329 overestimate AOD more than MERRA-2. In addition, in Northeast China, MERRA-2 has a
330 very good correlation ($r = 0.91$) and lower errors (RMSE = 0.07, MAE = 0.06) in the
331 summer than in the spring, winter, and autumn, while for CMIP6 the highest correlation (r
332 = 0.58) and lowest errors (RMSE = 0.12, MAE = 0.09) occur in the summer, followed by
333 spring, winter and autumn (Figs. 4 and 5). Over the North China Plain, MERRA-2 obtained
334 better AOD results in terms of r (0.86), RMSE (0.09), and MAE (0.07) during the autumn
335 than in the spring, winter, or summer, whereas in comparison the correlation ($r = 0.60$) and
336 errors (RMSE = 0.42, MAE = 0.38) for the CMIP6 AOD were comparatively higher in the
337 summer than in the spring, winter, or autumn. In the Yangtse River Delta, MERRA-2 AOD
338 compares best with the MODIS DTB AOD during the autumn as indicated by the high
339 correlation ($r = 0.81$) and the lower error metrics (RMSE = 0.11, and MAE = 0.10),
340 followed by summer, winter, and spring, whereas CMIP6 had inconsistent results. In

341 contrast, across Central China, consistent results between MERRA-2 AOD and MODIS
342 DTB AOD are obtained in the winter ($r = 0.82$, $RMSE = 0.14$, $MAE = 0.11$), followed by
343 summer, autumn, and spring, whereas overall CMIP6 had inconsistent results in all
344 seasons. However, for CMIP6 the correlation is higher and lower errors smaller in the
345 winter ($r = 0.61$, $RMSE = 0.18$, $MAE = 0.16$) than in the autumn, summer, or spring. In the
346 Sichuan Basin, MERRA-2 AOD and MODIS DTB AOD are well correlated in the summer
347 ($r = 0.77$, $RMSE = 0.21$, $MAE = 0.19$), but not in other seasons, while for CMIP6 the AOD
348 is not in good agreement with the MODIS DTB AOD in any season. In addition, both
349 MERRA-2 and CMIP6 significantly overestimate AOD during winter and autumn and
350 underestimates the AOD for the other two seasons (spring and summer), which contributes
351 to the inconsistent results over this area. Like for the North China Plain, the MERRA-2
352 AOD and MODIS DTB AOD across the Pearl River Delta region compare favorably in the
353 autumn ($r = 0.89$, $RMSE = 0.09$, $MAE = 0.07$) followed by spring, winter, and summer,
354 whereas for CMIP6 the AOD compare reasonably with the MODIS DTB AOD only for the
355 spring ($r = 0.79$, $RMSE = 0.18$, $MAE = 0.15$), rather than winter, autumn, or summer. Over
356 Qinghai, both MERRA-2 AOD and CMIP6 AOD are not in good agreement with the
357 MODIS DTB AOD for all seasons. Notably, CMIP6 overestimates the AOD in autumn and
358 summer and underestimates the AOD in the spring and winter, whereas MERRA-2
359 significantly underestimates the AOD in all seasons. The Tibetan Plateau is another low
360 AOD area in China, where MERRA-2 AOD is in good agreement with the MODIS DTB
361 AOD in the winter ($r = 0.81$, $RMSE = 0.09$, $MAE = 0.09$) followed by the spring, autumn,
362 and summer, whereas CMIP6 AOD compares well with the MODIS DTB AOD only in the
363 winter ($r = 0.79$, $RMSE = 0.04$, $MAE = 0.03$) and not for the other three seasons. In
364 addition, CMIP6 overestimates the AOD from spring to autumn and underestimates the
365 AOD in the winter, while MERRA-2 underestimates the AOD in all seasons. The Tarim

366 Basin is the biggest desert in China; here MERRA-2 AOD is in good agreement with the
367 MODIS DTB AOD in the summer ($r = 0.82$, $RMSE = 0.14$, $MAE = 0.13$) followed by the
368 autumn, winter, and spring, whereas CMIP6 AOD did not provide reasonable results in all
369 seasons. In comparison with MERRA-2, CMIP6 significantly overestimated the AOD in
370 the autumn, summer, and winter and underestimates the AOD in the winter. Across all
371 China, MERRA-2 AOD is in good agreement with the MODIS DTB AOD in the autumn (r
372 $= 0.86$, $RMSE = 0.02$, $MAE = 0.02$) followed by summer, winter, and spring, whereas
373 CMIP6 did not perform well in all seasons. CMIP6 significantly overestimates the AOD in
374 the autumn and winter and underestimates in the spring and summer, whereas MERRA-2
375 underestimates the AOD in all seasons. The underestimation of the AOD by MERRA-2 on
376 both annual and seasonal scales has been reported to probably be due to the uncertainty of
377 the emission inventory used in the GOES models (Buchard et al., 2017; Che et al., 2019b;
378 Shi et al., 2019). In contrast, the significant overestimation of the AOD by CMIP6 probably
379 results from the uncertainty of the Community Emissions Data System (CEDS) inventory
380 adopted by the CMIP6 models (Wang et al., 2021). Overall, MERRA-2 AOD performs
381 better over most regions of China than CMIP6, as concluded from the comparison with
382 MODIS DTB AOD data because of the high correlation and the low error metrics (RMSE
383 and MAE) for MERRA-2.

384 *3.2 Annual and seasonal mean spatial AOD patterns over China*

385 Figure 6 shows the spatial distributions of the annual and seasonal mean AOD,
386 obtained from CMIP6, MERRA-2, and MODIS DTB over China, averaged over the period
387 2000–2014. Between these three data sets, the annual mean MODIS DTB AOD is highest
388 (> 0.8) over the Central (Henan, Hubei, Hunan), East (Anhui, Jiangsu, Shanghai,
389 Shangdong), North (Tianjin and Hebei), and Southwest (the eastern part of the Sichuan

390 Basin and the western part of Chongqing) regions of China, whereas the CMIP6 and
391 MERRA-2 over these regions are lower (0.6 – 0.8) (Fig. 6). The second-highest mean
392 MODIS DTB AOD (0.6 – 0.8) is observed over parts of Anhui, Hunan, Jiangxi, Zhejiang,
393 Guandong, Guangxi, and Shanxi. Also in these regions, the CMIP6 and MERRA-2 mean
394 AOD is lower (0.5 – 0.6) than the MODIS DTB AOD. These areas are all low altitude
395 regions (< 500 m above sea level), characterized by high population density and high
396 anthropogenic aerosol emissions owing to rapid urbanization and industrialization (Luo et
397 al., 2014). Besides, Cao et al. (2014) reported that large amounts of coarse particles (soot
398 and dust) were emitted from industrial activities and coal fuel combustion, which constitute
399 a major contribution to the total aerosol loadings over these regions. The high AOD over
400 North China mainly results from the emission of coarse particles (desert dust) from the
401 Taklamakan Desert (Yu et al., 2016; Proestakis et al., 2018). Figure 6 shows that MODIS
402 DTB detected moderate levels of AOD (0.4 – 0.5) over parts of the Northeast (Liaoning,
403 Jilin, Heilongjiang), Fujian, Zhejiang, the Southwest province of Guizhou, the Northwest
404 province of Shanxi, and the Tarim Basin (Xinjiang), with the MERRA-2 AOD close to
405 MODIS DTB AOD and CMIP6 AOD higher than MODIS DTB AOD, especially over the
406 Tarim Basin (Xinjiang). The lowest AOD (< 0.30) was observed by MODIS DTB AOD
407 over high altitude areas with sparse populations such as the North (Inner Mongolia),
408 Northwest (Gansu, Qinghai, Ningxia), and West (Tibetan Plateau) of China. For these
409 regions, MERRA-2 again performed close to MODIS DTB AOD, but CMIP6
410 overestimated AOD (Fig. 6). Several previous studies also found the lowest AOD over
411 Inner Mongolia, Gansu, Qinghai, Ningxia, and the Tibetan Plateau (Liu et al., 2016; de
412 Leeuw et al., 2018). In conjunction with the above spatial information, seasonally, the
413 MODIS DTB AOD is highest in spring followed by summer, winter, and autumn
414 throughout China, where CMIP6 and MEERA-2 also show similar seasonal scenarios with

415 higher and lower AOD values than MODIS DTB AOD. It is likely that widespread
416 biomass-burning activities, less vegetation, and large amounts of sand and dust lifted from
417 their natural source (the Taklamakan and Gobi Desert) result in high AOD in spring. These
418 results are supported by previously published studies (Luo et al., 2014; He et al., 2016; Liu
419 et al., 2016). As more photochemical reactions occur during the summer, this might be
420 considered as another important possible factor for the second-highest aerosol loadings
421 over the study area (Dickerson et al., 1997). Li and Wang (2014) also documented that the
422 summertime AOD may be influenced by the abundant water vapor and droplets suspended
423 in the atmosphere during the summer season over China.

424

425 *3.3 Annual and Seasonal mean AOD variability using regional average*

426 Aerosol Optical Depth (AOD) obtained from CMIP6, MERRA-2, and MODIS DTB
427 was averaged from 2000 to 2014 at annual and seasonal scales over the ten major regions
428 selected across China (see Fig.1), for each of the years included in this study (2000–2014).
429 The results are plotted as time series as shown in Figs. 7 and Supplementary Data Figs. S1–
430 S4. The 15-year annual mean high AOD from MODIS DTB was 0.71 ± 0.08 in the North
431 China Plain, where comparatively lower AOD from CMIP6 (0.50 ± 0.04) and MERRA-2
432 (0.56 ± 0.07) was observed than MODIS DTB (Fig. 7). High AOD values probably result
433 from anthropogenic aerosols produced by industrial and vehicular emissions over these
434 regions, as was also reported by Hu et al. (2018). Due to the nature of the topography and
435 the sparse population, the lower annual mean AOD from MODIS DTB was 0.13 ± 0.01 in
436 the Tibetan Plateau, where CMIP6 (0.16 ± 0.01) and MERRA-2 (0.06 ± 0.005) also show
437 their lowest AOD than MODIS DTB AOD, signifying over- and –underestimation of AOD
438 (Fig. 7 and Table 2).

439 Seasonal patterns in the AOD were obvious over different regions of China
440 (Supplementary Data Figs. S1–S4 and Table 2). Seasonal mean MODIS DTB AOD was
441 highest in the spring in most regions of China except for North China Plain, where AOD
442 peaked in the summer than other seasons. In the spring, MODIS DTB AOD was highest in
443 Central China (0.81 ± 0.06), where the comparable result was observed from CMIP6 (0.81
444 ± 0.09) and MERRA-2 underestimates AOD (0.72 ± 0.09) compared to MODIS DTB AOD
445 (Table 2). It is important to mention that MODIS DTB AOD was lowest in the autumn over
446 the North China Plain, Yangtse River Delta, Tarim Basin, Northeast, Gobi Desert, Qinghai
447 as well as Entire China. In the autumn, MODIS DTB AOD was highest in the North China
448 Plain (0.57 ± 0.07), where both CMIP6 (0.48 ± 0.05) and MERRA-2 underestimate AOD
449 (0.50 ± 0.06) than MODIS DTB AOD (Table 2). Several earlier studies also found the
450 highest AOD in the spring and lowest in the autumn (Pan et al., 2010; Deng et al., 2012;
451 Luo et al., 2014; Cheng et al., 2015; Wang et al., 2015; He et al., 2016; De Leeuw et al.,
452 2018). Furthermore, across Central China, the Sichuan Basin, and the Tibetan Plateau of
453 China, MODIS DTB reported low seasonal mean AOD in the winter season. In the winter,
454 MODIS DTB AOD was highest in Central China (0.59 ± 0.08), where both CMIP6 ($0.75 \pm$
455 0.08) and MERRA-2 underestimate AOD (0.69 ± 0.11) than MODIS DTB AOD (Table 2).
456 Apart from the above-mentioned areas of China, across the Pearl River Delta, MODIS DTB
457 reported low AOD in the summer (0.46 ± 0.08), where both CMIP6 (0.38 ± 0.03) and
458 MERRA-2 underestimate AOD (0.32 ± 0.04) than MODIS DTB AOD (Table 2). Overall,
459 the study concludes that both CMIP6 and MERRA-2 substantially over- and under-
460 estimate AOD across China compared to MODIS DTB AOD, therefore, in section 3.4, the
461 study has calculated AOD trends using MODIS DTB AOD.

462

463 *3.4 Trends in AOD*

464 Figure 8 shows the spatial distributions of annual and seasonal trends of AOD
465 obtained from MODIS DTB over China for the period 2000 to 2014. It is evident from Fig.
466 8 and Table 3 that not all regions have statistically significant trends. However, a
467 substantial spatial contrast in AOD trends (both increasing and decreasing) was evident
468 over the study area at annual and seasonal scales (Fig. 8). Annually, decreasing AOD trends
469 from MODIS DTB were evident over the Gobi Desert ($-0.004/\text{year}$) and Qinghai
470 ($-0.001/\text{year}$). Guo et al. (2011) and Li (2020) also found a decreasing AOD trend over the
471 Gobi Desert. In addition, MODIS DTB reported increasing AOD trends from $0.0002/\text{year}$
472 to $0.014/\text{year}$ across the Northeast, North China plain, Yangtse River Delta, Central,
473 Sichuan Basin, Pearl River Delta, Tarim Basin, and Tibetan Plateau, as well as over Entire
474 China. The increasing AOD trends may be due to the rapid increase in industrial and
475 anthropogenic activities over these regions, resulting in abundant aerosols emitted into the
476 atmosphere; similar results were documented in Zhang et al. (2013) and Gui et al. (2017).
477 Wang et al. (2015) reported that sulfate aerosols from industry were mainly responsible for
478 the increasing AOD trends.

479
480 In the winter season, increasing AOD trends from MODIS DTB were observed which
481 varied between $0.0003/\text{year}$ and $0.020/\text{year}$ over most regions of China, except for the Gobi
482 Desert and Tibetan Plateau (Table 3). The increasing AOD trends during the winter may be
483 associated with meteorological conditions in China. Yin et al. (2017) reported that
484 meteorological conditions play a significant role in transporting pollutants and mixing
485 aerosols with local emissions from anthropogenic activities, resulting in an increase in
486 wintertime haze aerosols over China over the last few decades. Wind alone can explain

487 about 10% of the historical rise of regional aerosols (Gu et al., 2018). Liu et al. (2016)
488 documented that different anthropogenic activities linked with biomass and fossil fuel
489 burning were mainly responsible for enhancing AOD levels over Central China, East, and
490 Northeast during the autumn and winter, resulting in increasing AOD trends there. During
491 the spring, MODIS DTB AOD trends were decreasing with values varying from
492 0.00002/year to 0.009/year over the Gobi Desert, Northeast, Sichuan Basin, Qinghai, as
493 well as across Entire China (Table 3). The decreasing trends indicate a large decrease in
494 coarse-mode aerosol particles generated from natural sources in China. A similar result was
495 reported by Hu et al. (2018). Apart from this, the increasing AOD trends from MODIS
496 DTB from 0.001/year to 0.011/year were observed over the North China Plain, Yangtse
497 River Delta, Central, Pearl River Delta, Tibetan Plateau, and Tarim Basin (Table 3). The
498 increasing AOD trends over South China (Yangtse River Delta) during the spring were
499 attributed to the increased fine-mode particles from large anthropogenic emissions and
500 coarse-mode particles (sea-salt aerosols) from the coast of South China (Dong et al., 2013;
501 Qi et al., 2013; Luo et al., 2014). In the summer, the MODIS DTB AOD trends were
502 decreasing AOD by 0.002–0.007 (per year) over the Gobi Desert, Pearl River Delta,
503 Qinghai, and Tarim Basin (Table 3). Apart from this, the MODIS DTB AOD trends were
504 increasing from 0.0003/year to 0.022/year over the Northeast, North China Plain, Yangtse
505 River Delta, Central, Sichuan Basin, Tibetan Plateau, and Entire China (Table 3). Hu et al.
506 (2018) also found increasing AOD trends over the highly populated and industrialized
507 regions of Central, East, and Northeast of China. The increasing trends reflect the
508 dominance of fine-mode non-absorbing aerosols due to industrial emissions of SO₄ aerosol
509 (Wang et al., 2015). The decreasing AOD trend over the Pearl River Delta was also
510 documented by (Li, 2020). In the autumn, the MODIS DTB AOD trends were increasing
511 and varied from 0.0001/year to 0.010/year over the Northeast, North China Plain, Yangtse

512 River Delta, Central, Pearl River Delta, Qinghai, Tibetan Plateau, Tarim Basin, and Entire
513 China except for the Gobi Desert and Sichuan Basin, where trends were decreasing with
514 -0.0002 to -0.006 (per year) (Table 3).

515 In light of the above, AOD increased annually over most regions of China between
516 2000 and 2014, except for the Gobi Desert and Qinghai, where AOD was found to have
517 reduced. Seasonal deviations in AOD trends were also evident and are different between
518 different parts of China. The SO_2 and primary aerosol emissions increased substantially,
519 relative to dust from 2000 to 2005 due to rapid economic and industrial development,
520 which led to increased AOD (Zhao et al., 2017). The change (increase or decrease) in AOD
521 from 2000 to 2014 can be associated with changes in both meteorology and emissions (Li,
522 2020). In the next section, we further investigate if there were any co-benefits from China's
523 strict air pollution control policies on AOD during the 11th and 12th Five-Year Plan (FYP)
524 periods. Therefore, we calculated AOD linear trends for 2000–2005, 2006–2010 (11th
525 FYP), and 2011–2014 (12th FYP) (Figs. 9–12).

526 MODIS DTB AOD trends over China were noticeably decreasing at annual and
527 seasonal scales during the 12th FYP period, following the 11th FYP period and 2000–2005,
528 which was probably due to the implementation of strict air pollution control policies (Figs.
529 9–11). On the annual scale, MODIS DTB AOD trends were strongly decreasing with
530 $-0.003/\text{year}$ to $-0.074/\text{year}$ during the 12th FYP period throughout the whole country,
531 except for Northeast, Qinghai, and Tibetan Plateau. These decreasing trends were stronger
532 during the 12th FYP period than during the 11th FYP period and 2000–2005 (Fig. 12). On a
533 seasonal scale, MODIS DTB AOD also strongly decreased during the 12th FYP period
534 across different parts of China as compared to the 11th FYP period and 2000–2005 (Fig.
535 12). Although China's air pollution control policies during the 11th and 12th FYP periods

536 were not designed for AOD control and prevention, they still had co-benefits as AOD was
537 subsequently found to decrease at both annual and seasonal scales over China.

538 Several reasons might be considered for the increase and decrease in AOD over the
539 study area. For example, due to rapid economic and industrial development from 2000 to
540 2005, the emissions of SO₂ and primary aerosols increased substantially relative to dust,
541 resulting in increased AOD over China (Zhao et al., 2017). The reduction of SO₂ and PM_{2.5}
542 (contains: sulfate, nitrate, organic carbon, elemental carbon), and the control of industrial
543 dust with soot (Zhou et al., 2015; Jin et al., 2016; Ma et al., 2019) may have contributed to
544 a decrease in AOD during the 11th FYP period. The effective reduction in anthropogenic
545 emissions could lead to AOD reduction (Sogacheva et al., 2018). The Environmental
546 protection, Energy Conservation and Emissions Reduction (ECER), and Air Pollution
547 Prevention and Control Key Regions (APPC-KR) policies during the 12th FYP period
548 reduced SO₂, NO_x, PM_{2.5}, PM₁₀, industrial dust, and soot emissions (Wang et al., 2018),
549 which contributed to decreasing AOD over China. Liu et al. (2021) reported that
550 anthropogenic aerosol species (black carbon, organic carbon, and sulfate) were reduced
551 over the Sichuan Basin because of strict implementation of the air pollution control policies
552 in China during the 12th FYP period, resulting in AOD also reduced in this regions.

553

554

555 **4. Conclusion**

556 In the current study, CMIP6 and MERRA-2 AOD were evaluated against MODIS DTB
557 AOD for the years 2000–2014, its long-term spatiotemporal variations were scrutinized
558 and the effectiveness of air pollution control policies to control AOD over China from
559 2000 to 2014 was considered.

- 560 • The evaluation results show higher correlation ($r = 0.71\text{--}0.89$) and lower error
561 (RMSE = $0.07\text{--}0.23$, MAE = $0.05\text{--}0.17$) for MERRA-2 than for CMIP6 ($r = 0.38\text{--}$
562 0.66 , RMSE = $0.05\text{--}0.28$, MAE = $0.04\text{--}0.23$) over the Gobi Desert, Northeast,
563 North China plain, Pearl River Delta, Tibetan Plateau, and Entire China. Likewise,
564 over the Yangtse River Delta, Central, Qinghai, and Tarim Basin, MERRA-2
565 correlation and error ($r = 0.51\text{--}0.64$, RMSE = $0.11\text{--}0.20$, MAE = $0.08\text{--}0.15$) are
566 better than that of CMIP6 ($0.06\text{--}0.45$, $0.13\text{--}0.36$, $0.09\text{--}0.29$). Over the Sichuan
567 Basin, poor comparison results were obtained for both MERRA-2 (0.19 , 0.23 , 0.19)
568 and the CMIP6 (-0.02 , 0.24 , 0.18). MERRA-2 underestimated AOD, which might
569 result from the uncertainty of the emission inventory used in the Goddard Earth
570 Observing System (GEOS) models. CMIP6 overestimated AOD over most regions,
571 possibly due to the uncertainty of the Community Emissions Data System (CEDS)
572 inventory adopted by CMIP6 models. In any case, it is clear that the CMIP6 and
573 MERRA-2 both need significant improvement in how they simulate AOD.
- 574 • During the study period from 2000 to 2014, AOD from Terra-MODIS DTB was
575 high across the low altitude regions characterized by highly populated,
576 economically, and industrially developed regions of the North China Plain ($0.71 \pm$
577 0.08) followed by Central China (0.69 ± 0.06), Yangtse River Delta (0.67 ± 0.05),
578 Sichuan Basin (0.64 ± 0.06), and Pearl River Delta (0.54 ± 0.05). AOD levels were
579 lowest over the Tibetan Plateau (0.13 ± 0.01) followed by Qinghai (0.19 ± 0.03) and
580 the Gobi Desert (0.21 ± 0.03), due to the role of topography and sparse population.
581 Seasonally, the highest AOD over China occurred in spring, followed by summer,
582 winter, and autumn. Widespread biomass-burning activities, less vegetation, and the
583 lifting of large amounts of sand and dust particles from natural sources (the
584 Taklamakan and Gobi deserts) may result in high AOD in spring. Moreover,

585 increased photochemical reactivity, and abundant water vapor and droplets in the
586 atmosphere may all contribute to the secondary AOD maximum during the summer.

587 • AOD trends were shown by Terra-MODIS DTB to be increasing throughout most
588 of the country during 2000–2014 and 2000–2005. Uncontrolled industrialization,
589 urbanization, and strong economic development mostly occurred from 2000 to 2005
590 in China, which may have led to the overall increasing AOD trends. AOD levels
591 decreased substantially from $-0.003/\text{year}$ to $-0.074/\text{year}$, throughout the whole
592 country except for the Northeast, Qinghai, and Tibetan Plateau (here AOD
593 increased by $0.001/\text{year}$ to $0.002/\text{year}$); these trends were greater than during the
594 11th FYP period. Seasonally, Terra-MODIS DTB AOD showed a prominent
595 decreasing trend in the summer season during the 12th FYP period across different
596 parts of China; stronger than during the 11th FYP period. During the 12th FYP
597 period, strict implementation of China's air pollution control policies may have
598 reduced the anthropogenic emissions of primary aerosols, SO_2 , NO_x , $\text{PM}_{2.5}$ (a
599 combination of sulfate, nitrate, organic carbon, elemental carbon), industrial dust,
600 and soot, which may be possible reasons for the AOD reduction. Evidently, the air
601 pollution control policies had the co-benefit of reducing AOD, resulting in
602 improved air quality over China. Overall, the size of the errors found in CMIP6 and
603 MERRA-2 AOD output suggests that they still cannot be effectively used for air
604 quality monitoring at regional and local scales within a country like China.

605

606 **CRedit authorship contribution statement**

607 **Md. Arfan Ali:** Conceptualization, Data curation, Methodology, Formal analysis,
608 Investigation, Validation, Visualization, Writing - original draft. **Muhammad Bilal:**
609 Conceptualization, Supervision, Investigation, Visualization, Writing - review & editing.

610 **Yu Wang:** Data curation, Methodology, Formal analysis, Investigation, Validation,
611 Visualization. **Zhongfeng Qiu:** Supervision, Writing - review & editing. **Janet E. Nichol,**
612 **Gerrit de Leeuw, Alaa Mhawish, Mansour Almazroui, M. Nazrul Islam, Usman**
613 **Mazhar:** Writing - review & editing. **Birhanu Asmerom Habtemicheal:** Formal analysis
614 & Visualization, **Song Ke:** Data curation.

615

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628 Supplementary data to this article can be found online at XXXXX

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630

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 1182 for major areas of China during the years 2000–2014.

1183 **Table 1** Summary of the 15 CMIP6 models used in the study.

S.L.	Models	Modelling Country	Variant Label	Spatial Resolution (°)
1	AWI-ESM-1-1-LR	Germany	rlilp1fl	0.9×0.9
2	BCC-ESM1	China	rlilp1fl	1.125×1.125
3	CanESM5	Canada	rlilp1fl	2.8×2.8
4	CESM2	USA	rlilp1fl	0.9×1.3
5	CESM2-FV2	USA	rlilp1fl	2.5×1.9
6	CESM2-WACCM	USA	rlilp1fl	1.3×0.9
7	CESM2-WACCM-FV2	USA	rlilp1fl	2.5×1.9
8	CMCC-CM2-SR5	Italy	rlilp1fl	1.0×1.0
9	E3SM-1-0	USA	rlilp1fl	1.0×1.0
10	GISS-E2-1-G	USA	rlilp5fl	2.0×2.5
11	HadGEM3-GC31-LL	UK	rlilp1f3	1.25×1.85
12	MPI_ESM_1_2_HAM	Germany	rlilp1fl	1.875×1.875
13	MPI_ESM1_2_HR	Germany	r5ilp1fl	1.0×1.0
14	NorESM2-LM	Norway	rlilp1fl	2.0×2.0
15	UKESM1-0-LL	UK	rlilp1fl	1.9×1.3

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1186 **Table 2** Mean annual and seasonal AOD obtained from CMIP6 Ensemble, MERRA-2
 1187 reanalysis, and Terra-MODIS over major areas of China for the period 2000–2014.

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Areas	AOD	Winter	Spring	Summer	Autumn	Annual
Gobi Desert	CMIP6	0.22±0.02	0.40±0.02	0.33±0.02	0.33±0.02	0.32±0.01
	MERRA-2	0.16±0.02	0.29±0.03	0.28±0.03	0.19±0.02	0.23±0.01
	MODIS DTB	0.15±0.02	0.31±0.06	0.24±0.05	0.13±0.02	0.21±0.03
Northeast	CMIP6	0.20±0.01	0.34±0.05	0.35±0.03	0.27±0.02	0.29±0.02
	MERRA-2	0.16±0.03	0.39±0.11	0.38±0.08	0.23±0.04	0.29±0.09
	MODIS DTB	0.24±0.06	0.42±0.11	0.34±0.08	0.23±0.06	0.31±0.08
North China Plain	CMIP6	0.40±0.04	0.55±0.04	0.58±0.05	0.48±0.05	0.50±0.04
	MERRA-2	0.41±0.07	0.58±0.09	0.73±0.11	0.50±0.06	0.56±0.07
	MODIS DTB	0.60±0.11	0.72±0.09	0.96±0.17	0.57±0.07	0.71±0.08
Yangtse River Delta	CMIP6	0.55±0.05	0.66±0.06	0.51±0.05	0.52±0.04	0.56±0.05
	MERRA-2	0.55±0.09	0.63±0.06	0.52±0.08	0.47±0.08	0.54±0.07
	MODIS DTB	0.61±0.07	0.77±0.05	0.74±0.10	0.56±0.06	0.67±0.05
Central	CMIP6	0.75±0.08	0.81±0.09	0.54±0.06	0.64±0.06	0.68±0.07
	MERRA-2	0.69±0.11	0.72±0.09	0.52±0.08	0.62±0.10	0.64±0.09
	MODIS DTB	0.59±0.08	0.81±0.06	0.73±0.12	0.64±0.09	0.69±0.06
Sichuan Basin	CMIP6	0.70±0.07	0.71±0.08	0.51±0.06	0.66±0.09	0.65±0.08
	MERRA-2	0.74±0.12	0.71±0.10	0.48±0.07	0.63±0.10	0.64±0.09
	MODIS DTB	0.48±0.17	0.80±0.07	0.67±0.08	0.60±0.08	0.64±0.06
Pearl River Delta	CMIP6	0.50±0.04	0.58±0.06	0.38±0.03	0.46±0.04	0.48±0.04
	MERRA-2	0.49±0.08	0.64±0.12	0.32±0.04	0.46±0.09	0.48±0.07
	MODIS DTB	0.48±0.07	0.71±0.07	0.46±0.08	0.52±0.09	0.54±0.05
Qinghai	CMIP6	0.14±0.01	0.24±0.01	0.23±0.02	0.25±0.03	0.22±0.01
	MERRA-2	0.06±0.01	0.18±0.01	0.14±0.051	0.08±0.01	0.12±0.01
	MODIS DTB	0.16±0.03	0.31±0.06	0.16±0.06	0.12±0.02	0.19±0.03
Tibetan Plateau	CMIP6	0.12±0.01	0.16±0.01	0.18±0.02	0.15±0.02	0.16±0.01
	MERRA-2	0.03±0.004	0.10±0.01	0.08±0.01	0.04±0.005	0.06±0.005
	MODIS DTB	0.09±0.01	0.20±0.02	0.13±0.02	0.10±0.01	0.13±0.01
Tarim Basin	CMIP6	0.31±0.04	0.68±0.04	0.69±0.06	0.68±0.03	0.59±0.03
	MERRA-2	0.21±0.03	0.47±0.04	0.48±0.05	0.34±0.05	0.37±0.02
	MODIS DTB	0.20±0.05	0.73±0.12	0.36±0.06	0.19±0.05	0.37±0.04
Entire China	CMIP6	0.30±0.03	0.43±0.03	0.38±0.02	0.38±0.03	0.37±0.02
	MERRA-2	0.24±0.03	0.37±0.04	0.32±0.03	0.26±0.03	0.30±0.03
	MODIS DTB	0.31±0.03	0.47±0.04	0.40±0.04	0.27±0.02	0.36±0.02

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1191 **Table 3** Trends in AOD derived from the MODIS DTB AOD data over major areas of China

1192 from 2000–2014. The asterisk (*) indicates significance at 95% confidence level.

Areas	Winter	Spring	Summer	Autumn	Annual
Gobi Desert	-0.002	-0.006	-0.007*	-0.0002	-0.004*
Northeast	0.008*	-0.009	0.007	0.008*	0.003
North China Plain	0.020*	0.005	0.022*	0.010*	0.014*
Yangtse River Delta	0.012*	0.001	0.008	0.005	0.006*
Central China	0.014*	0.003	0.011	0.005	0.008*
Sichuan Basin	0.008	-0.0003	0.006	-0.006	0.001
Pearl River Delta	0.003	0.011*	-0.002	0.0001	0.003
Qinghai	0.0003	-0.00002	-0.005	0.001	-0.001
Tibetan Plateau	-0.0002	0.001	0.0003	0.0002	0.0002
Tarim Basin	0.0004	0.003	-0.004	0.006*	0.001
Entire China	0.002	-0.0003	0.002	0.003*	0.002

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1195 **Declaration of interests**

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1197 The authors declare that they have no known competing financial interests or personal
 1198 relationships that could have appeared to influence the work reported in this paper.

1199

1200 The authors declare the following financial interests/personal relationships which may be
 1201 considered as potential competing interests:

1202

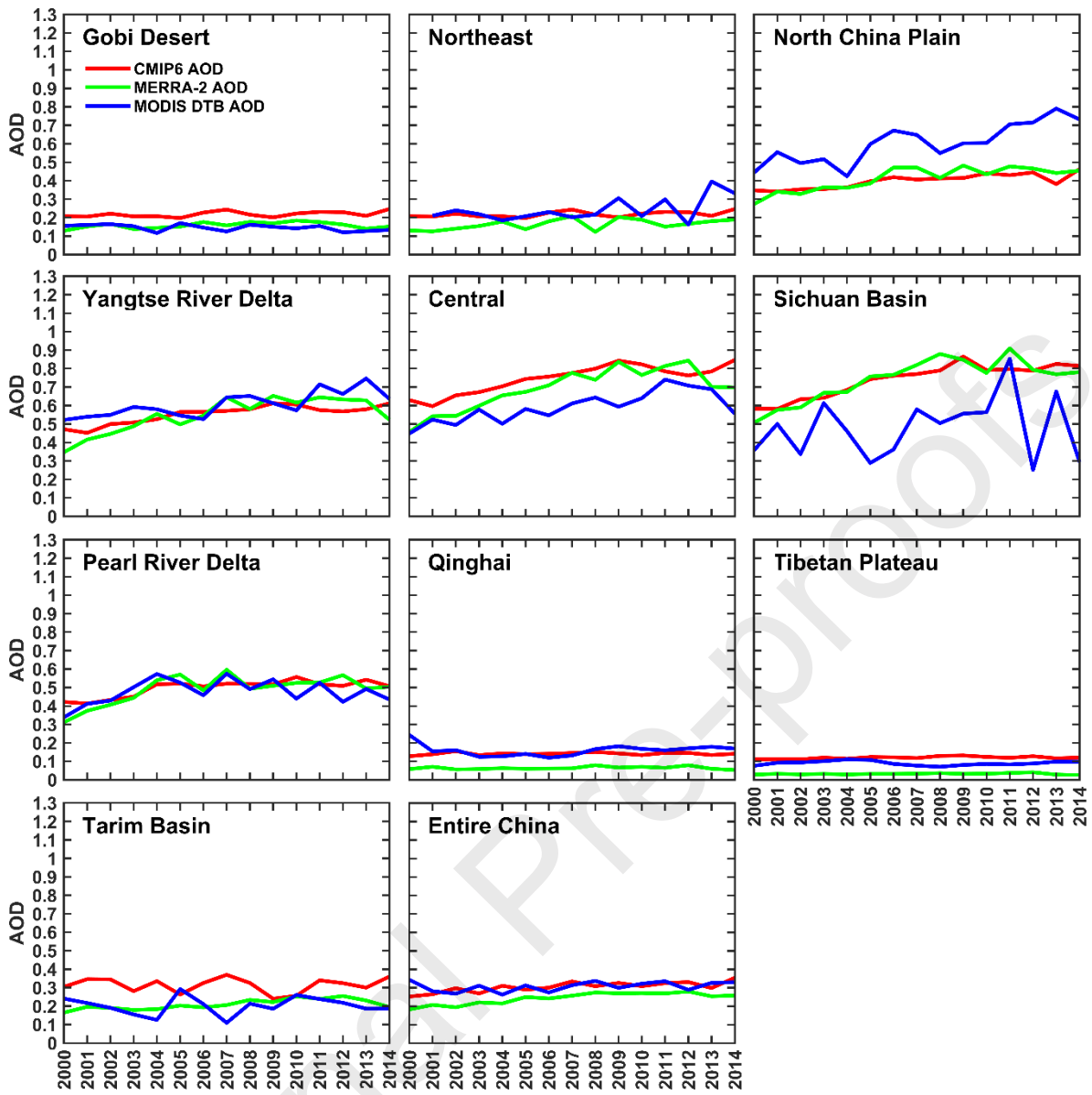
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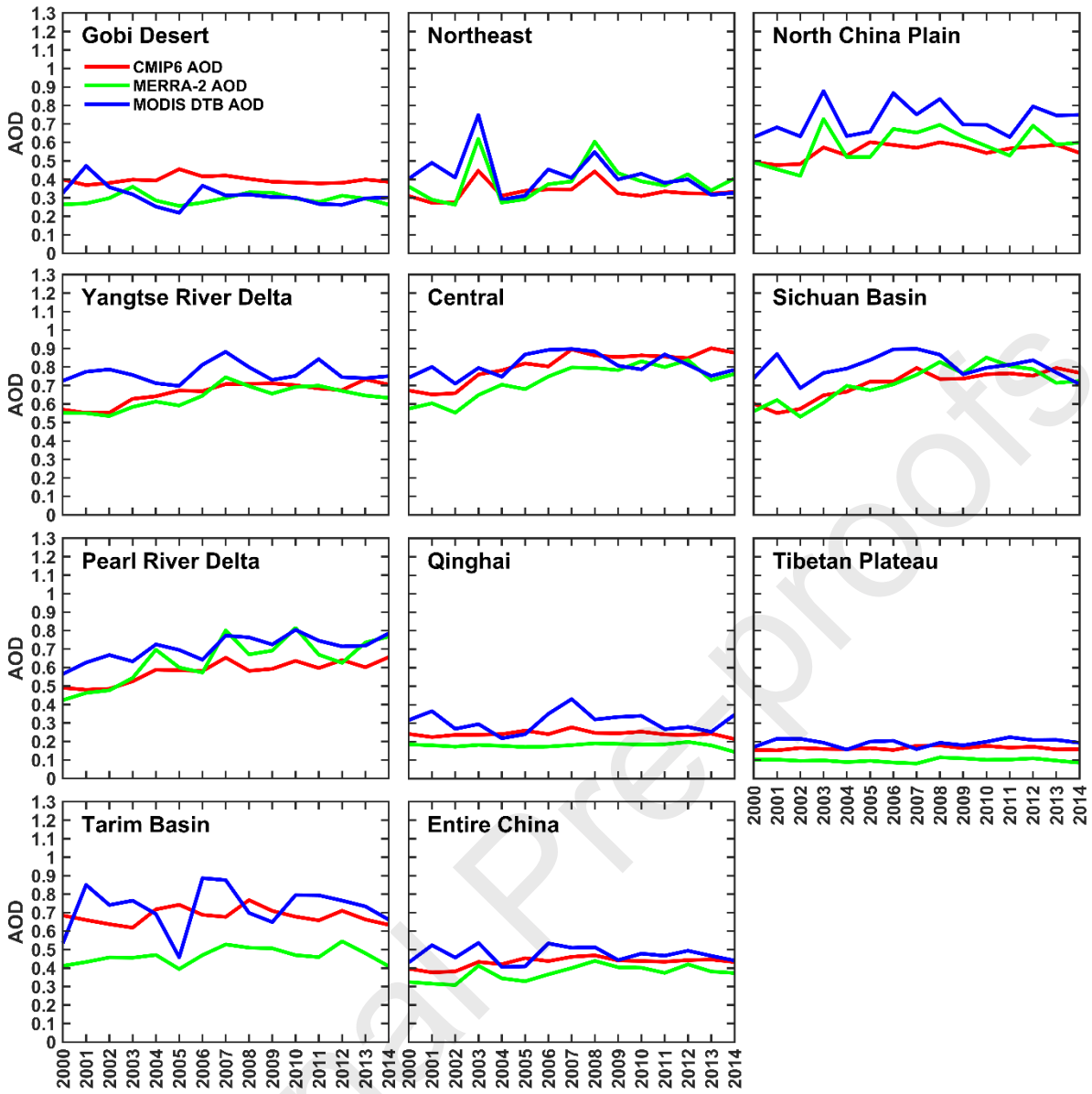
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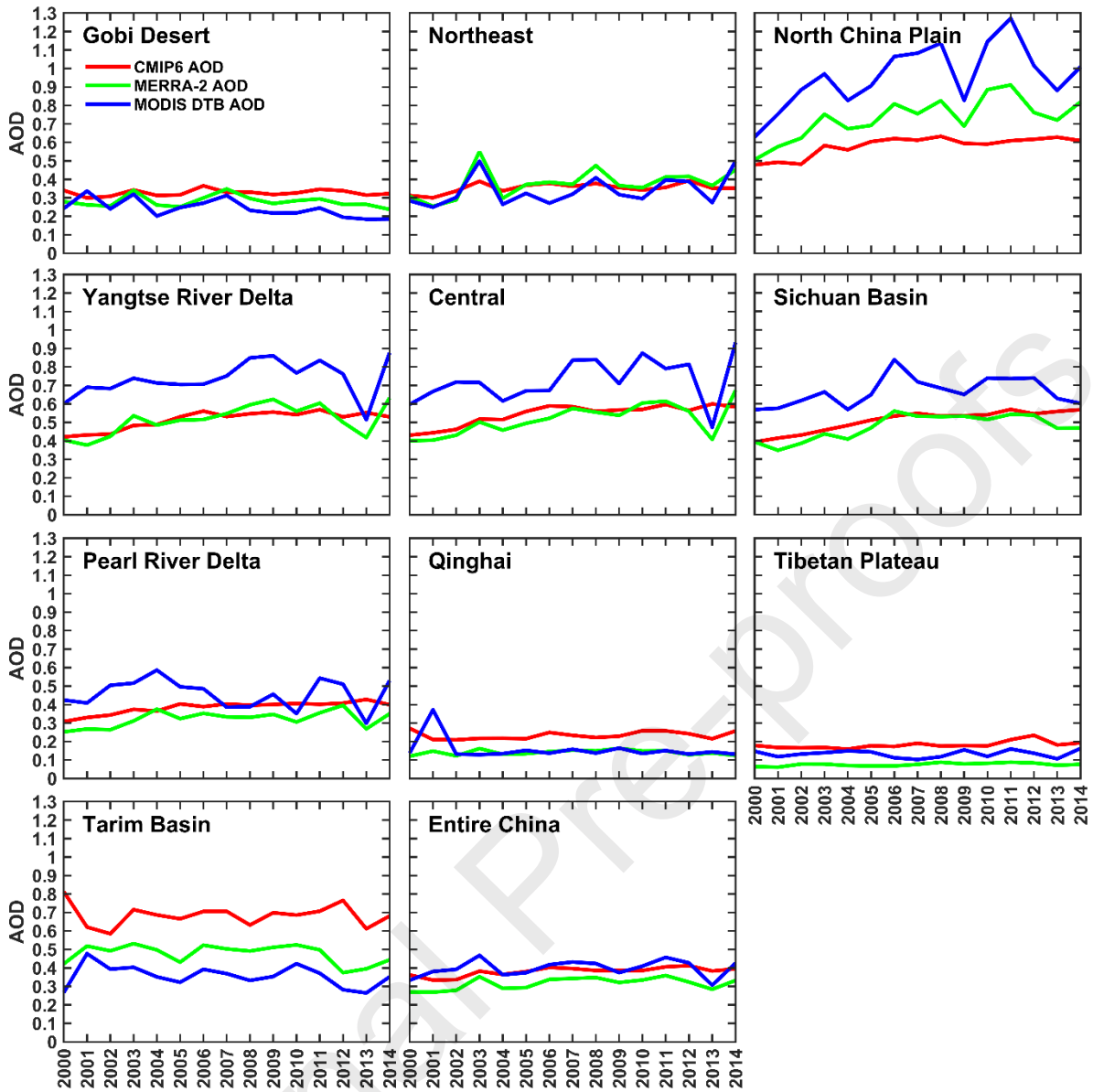
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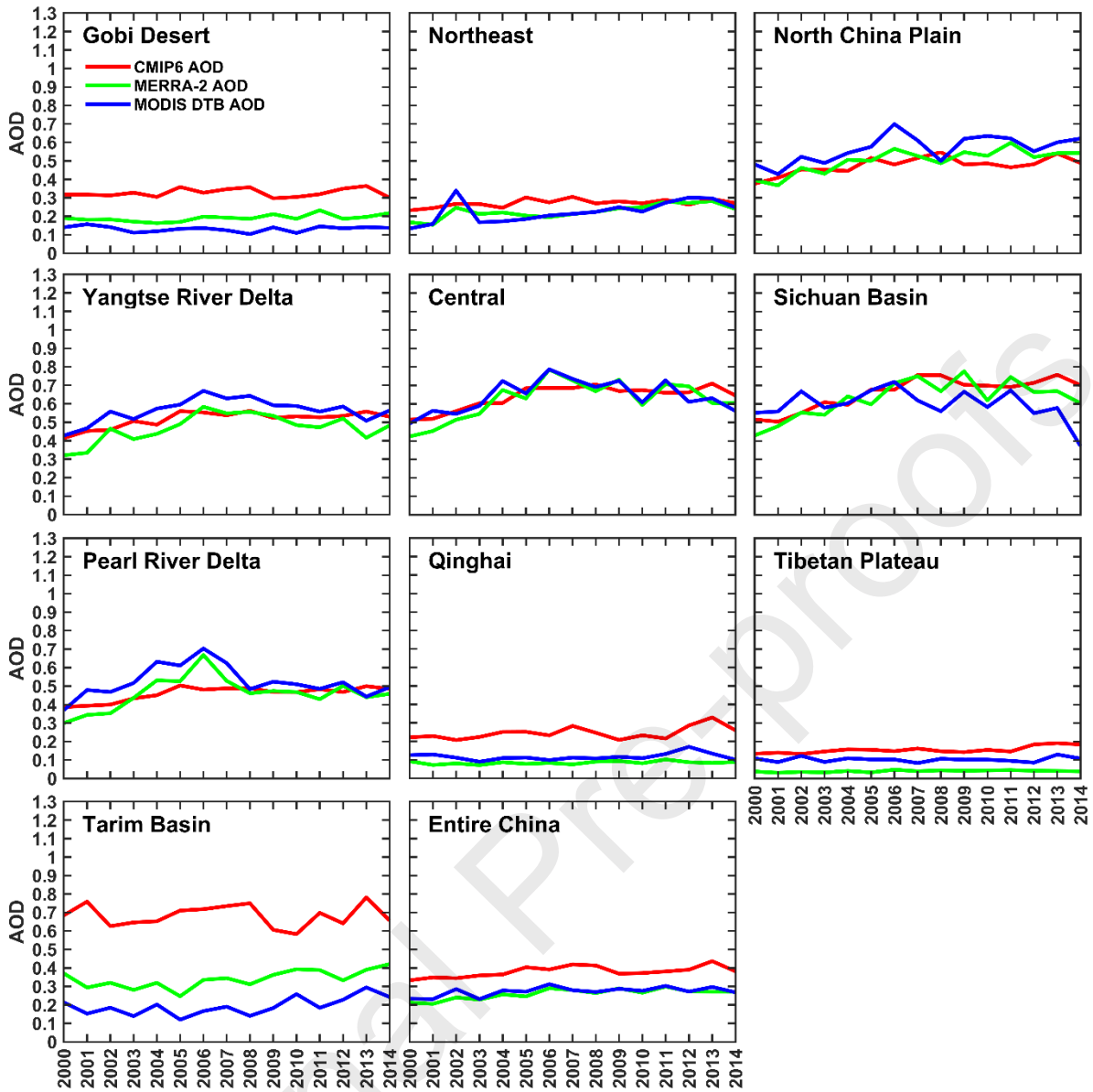
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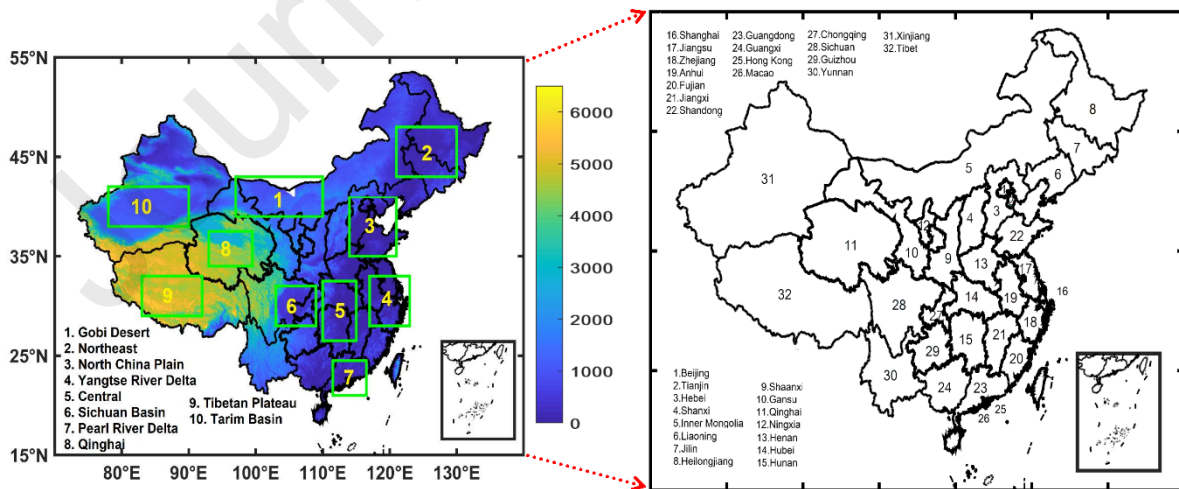
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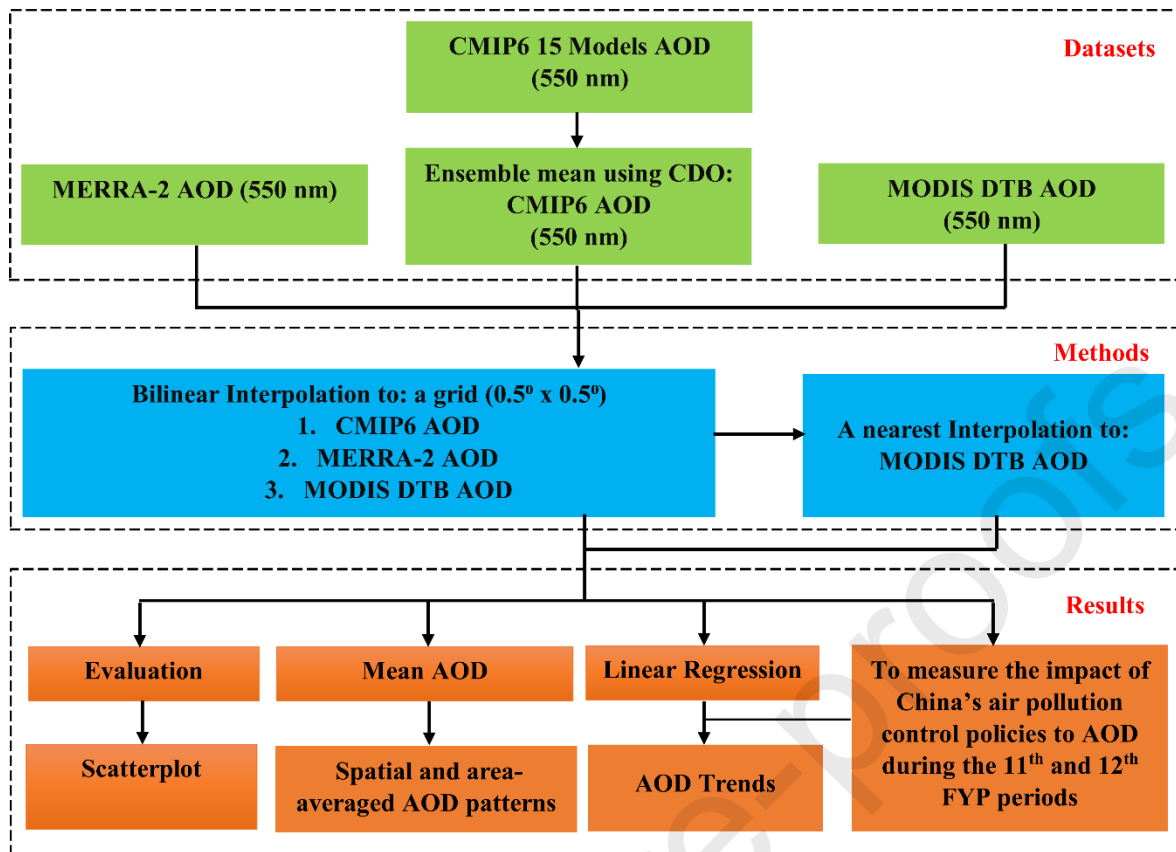
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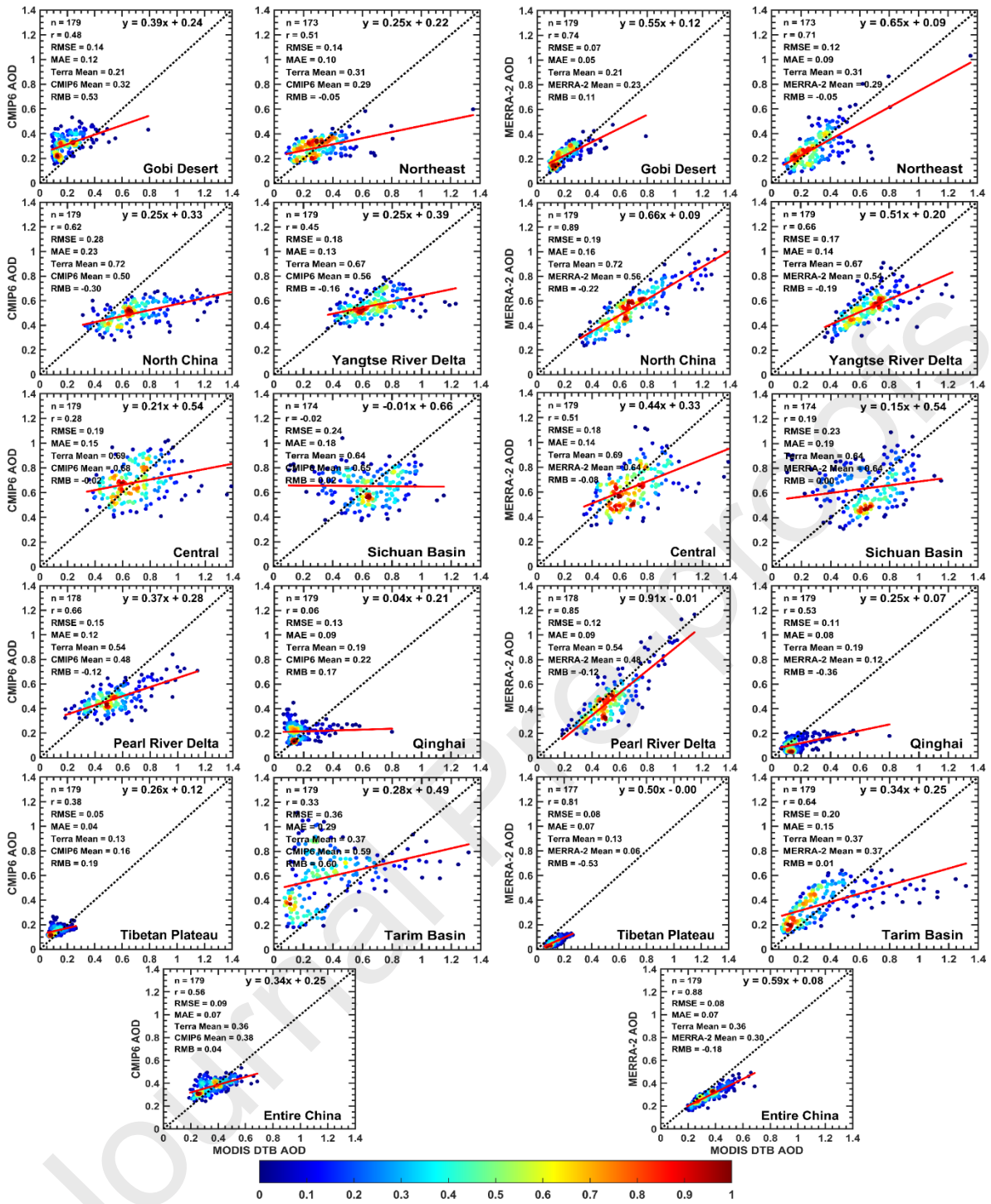
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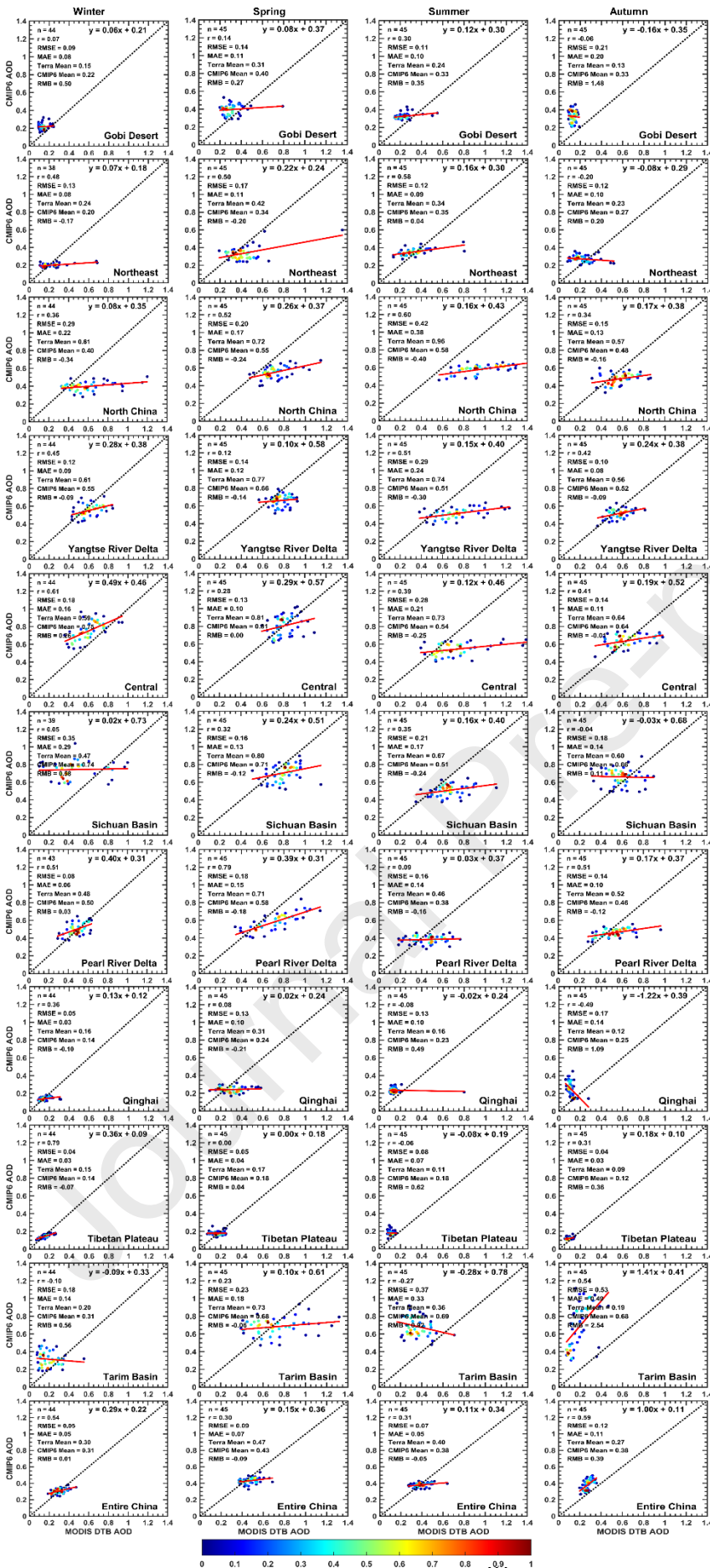
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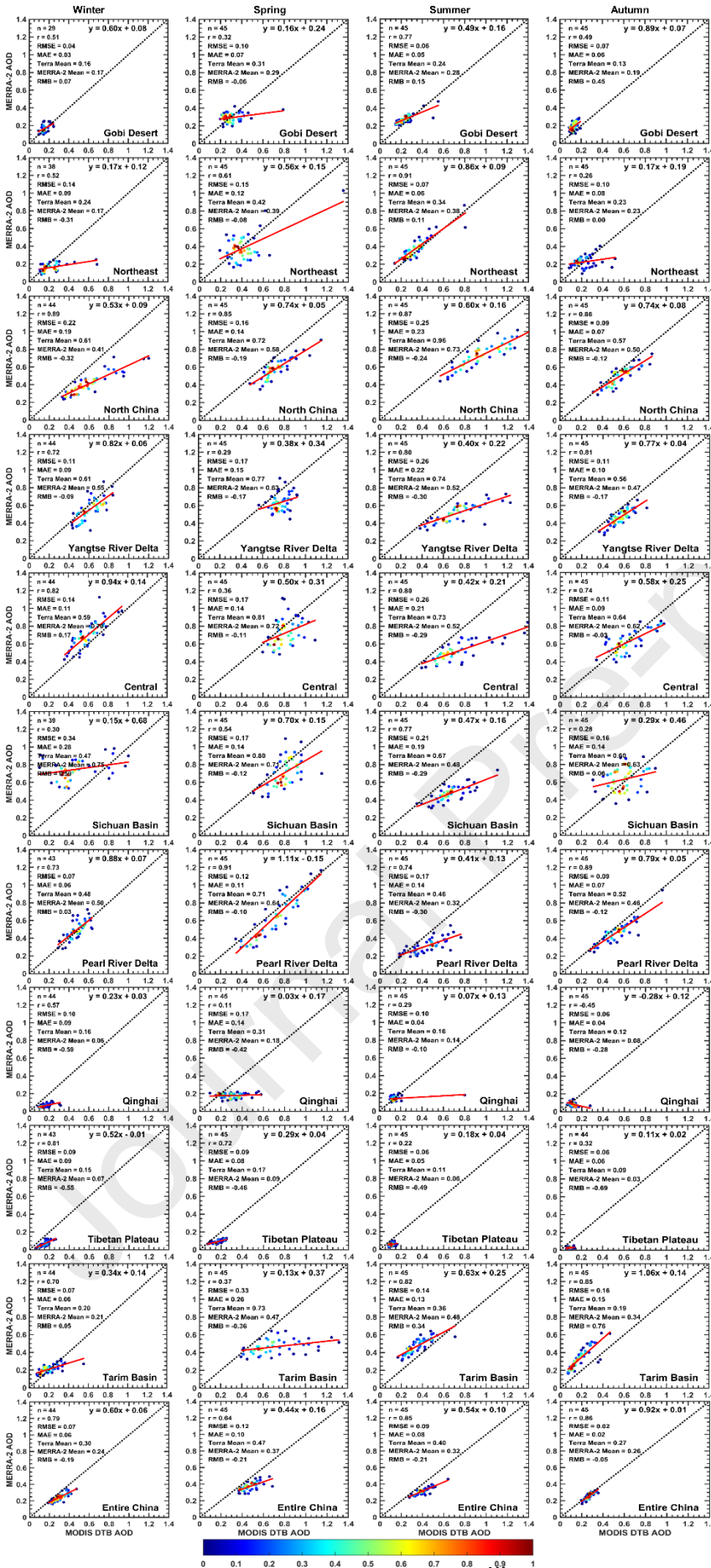


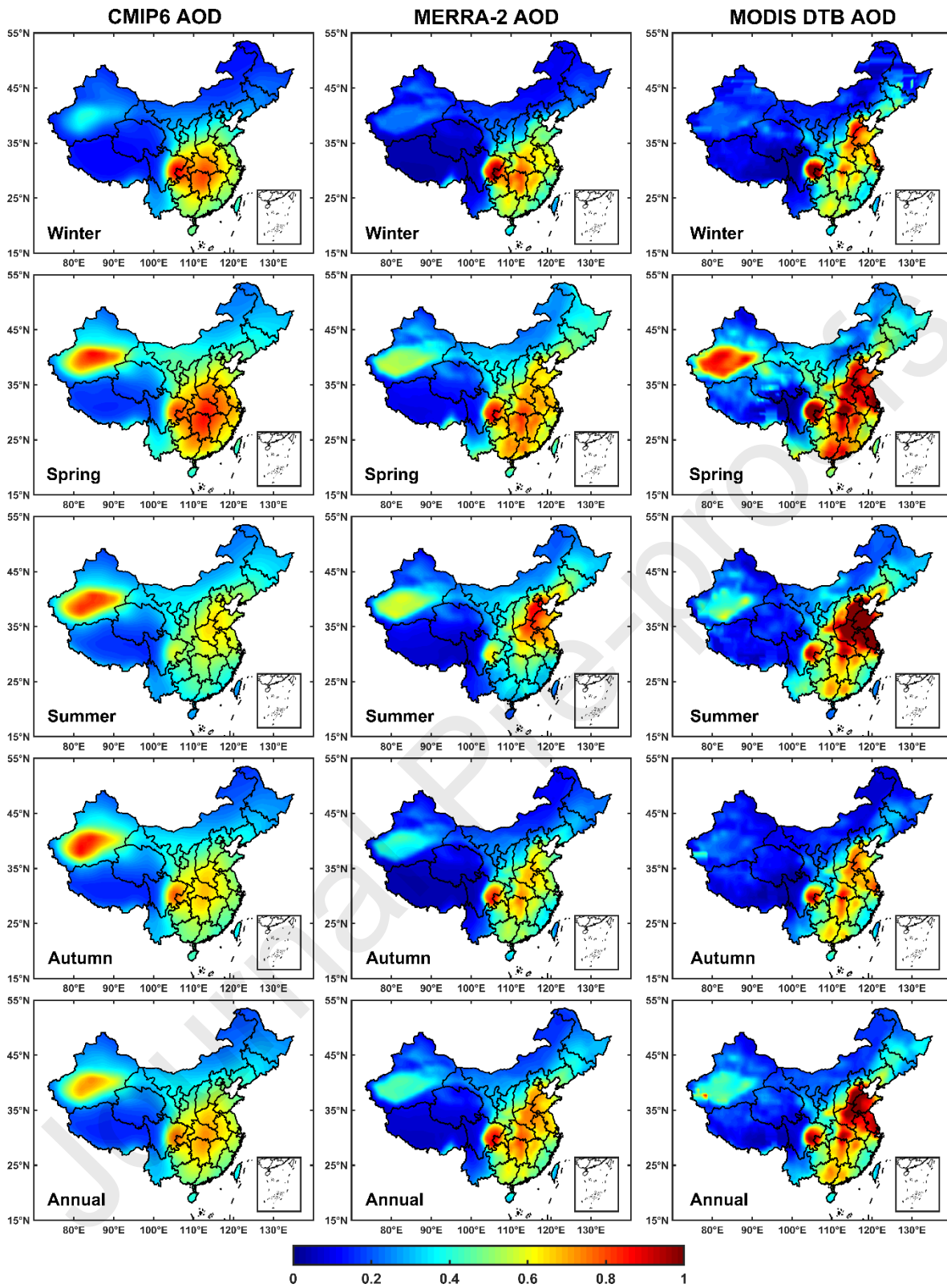
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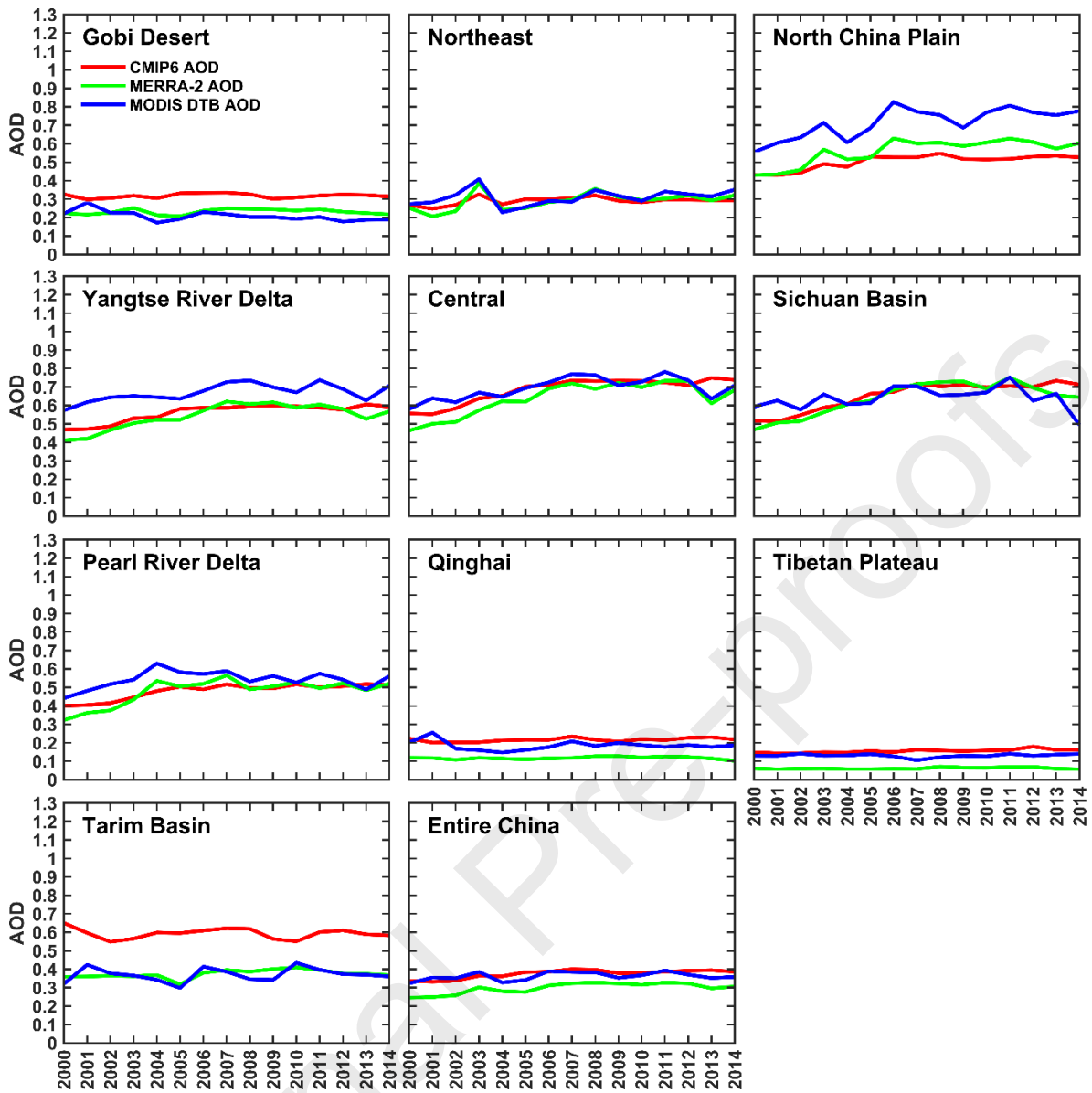
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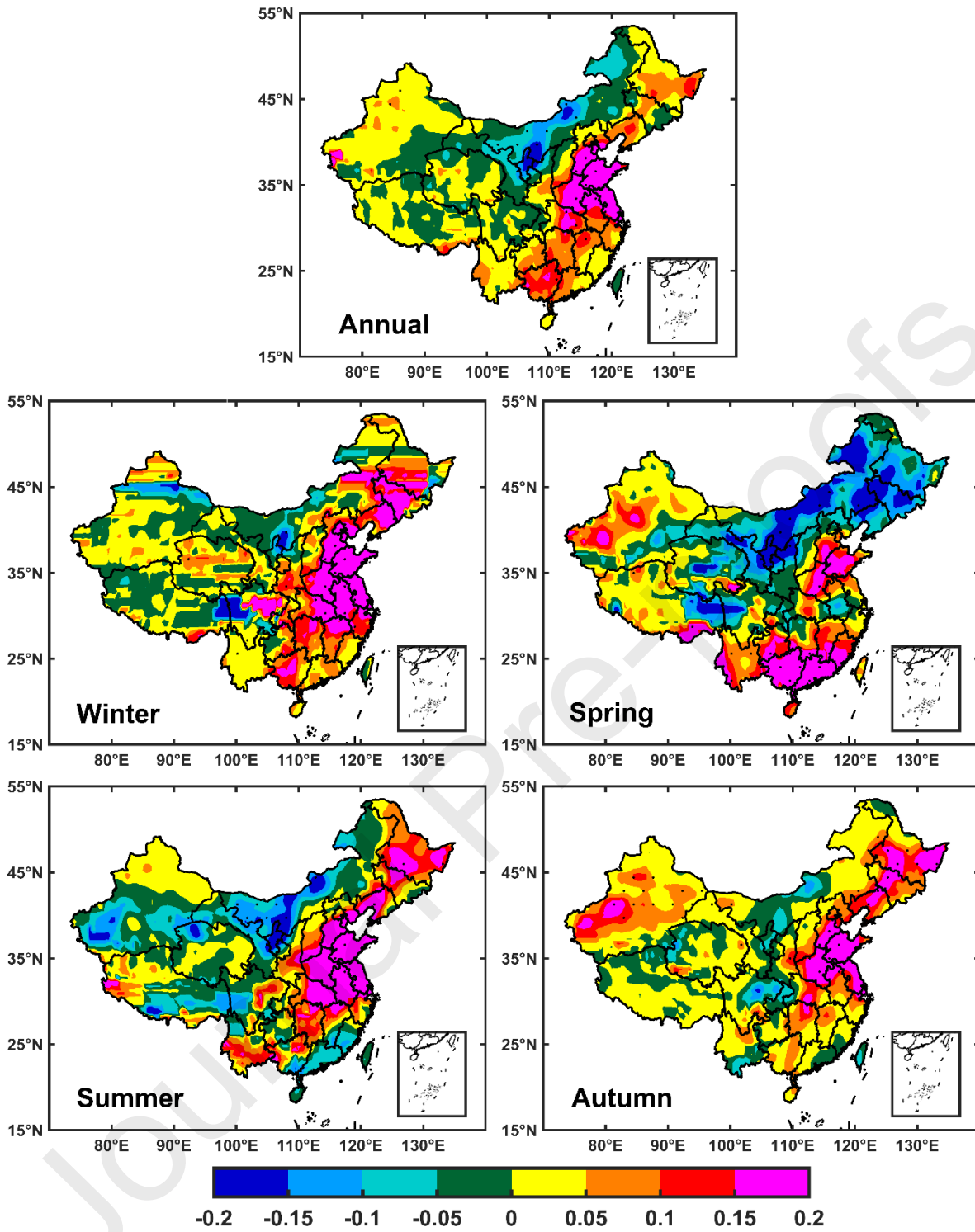




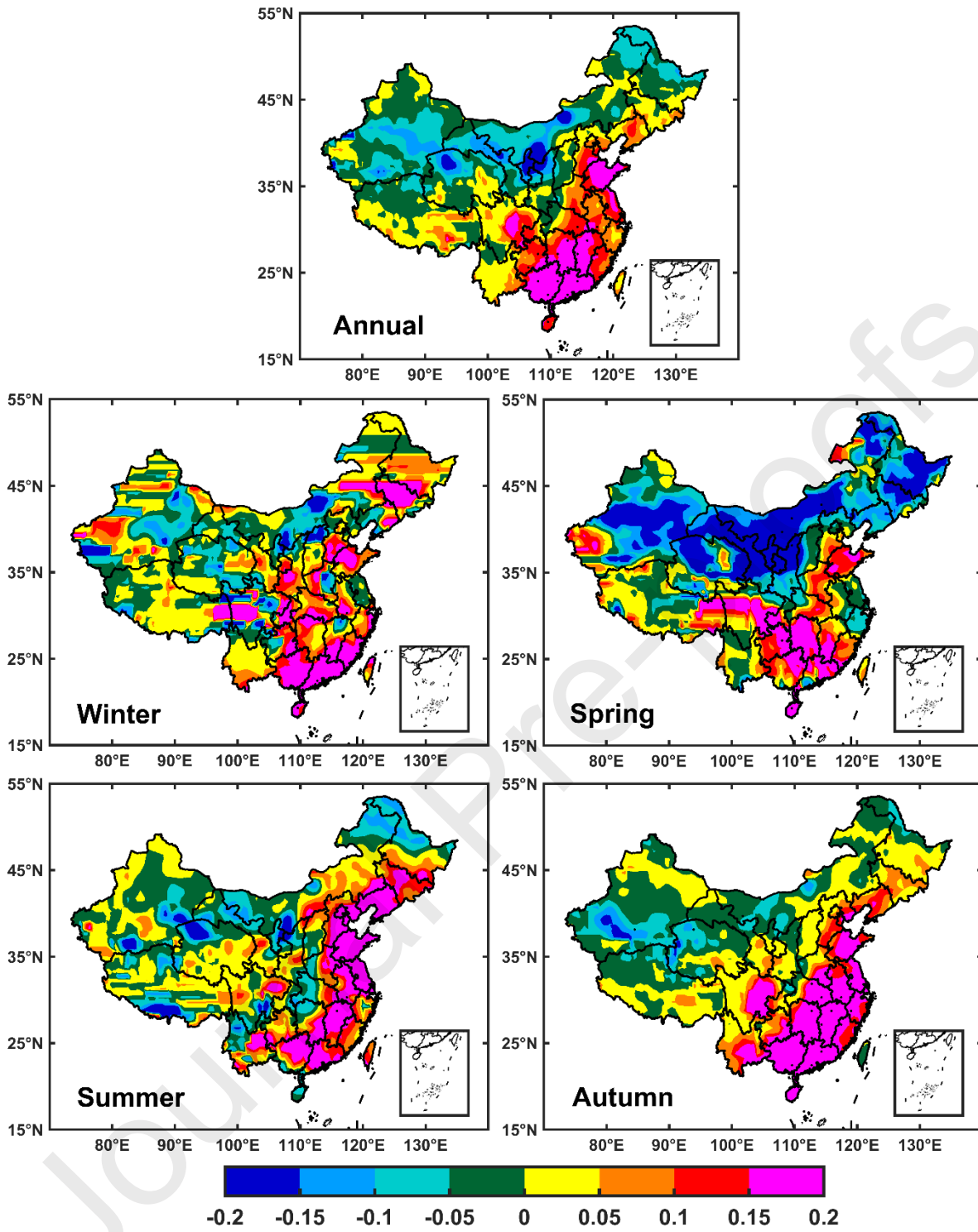
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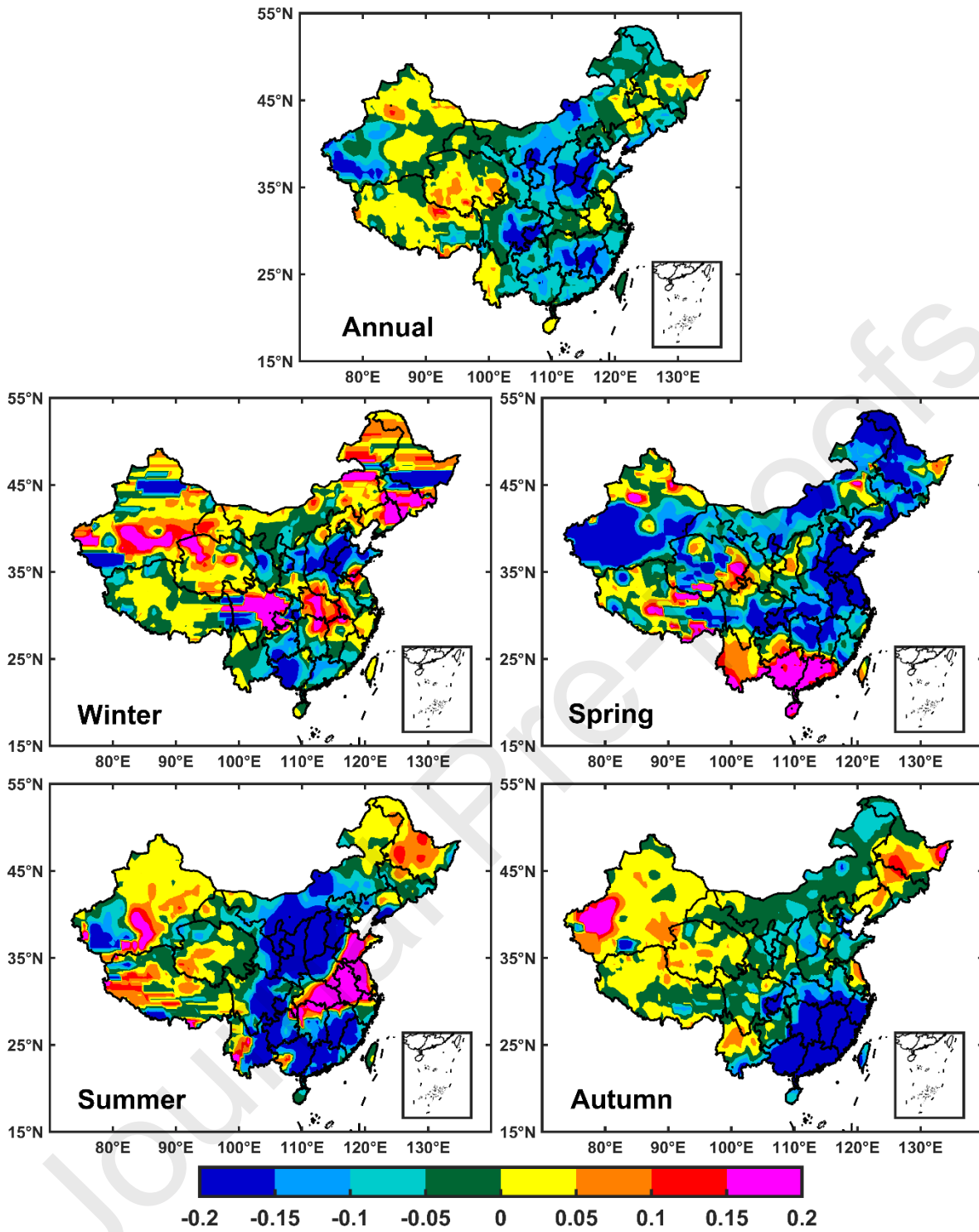
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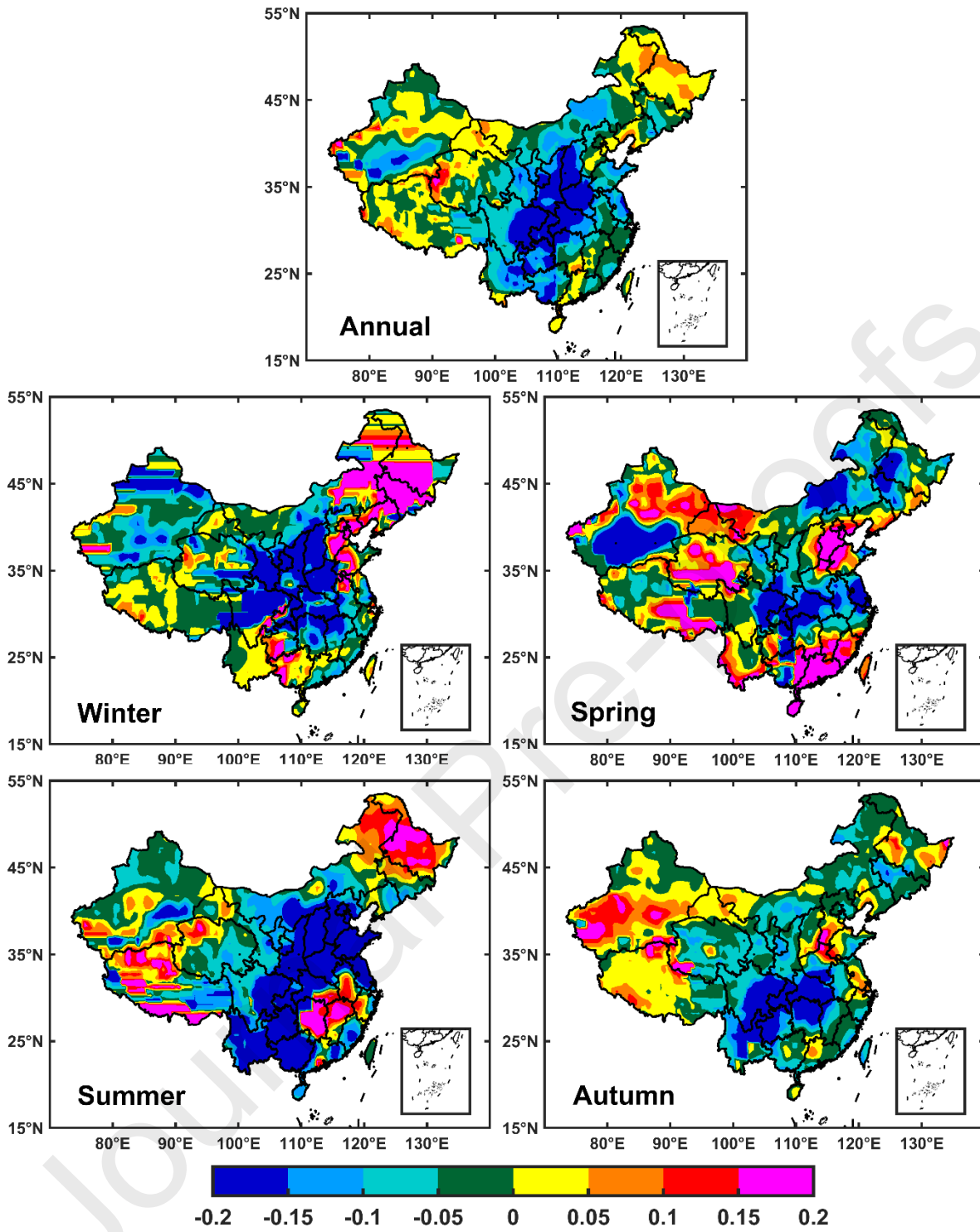
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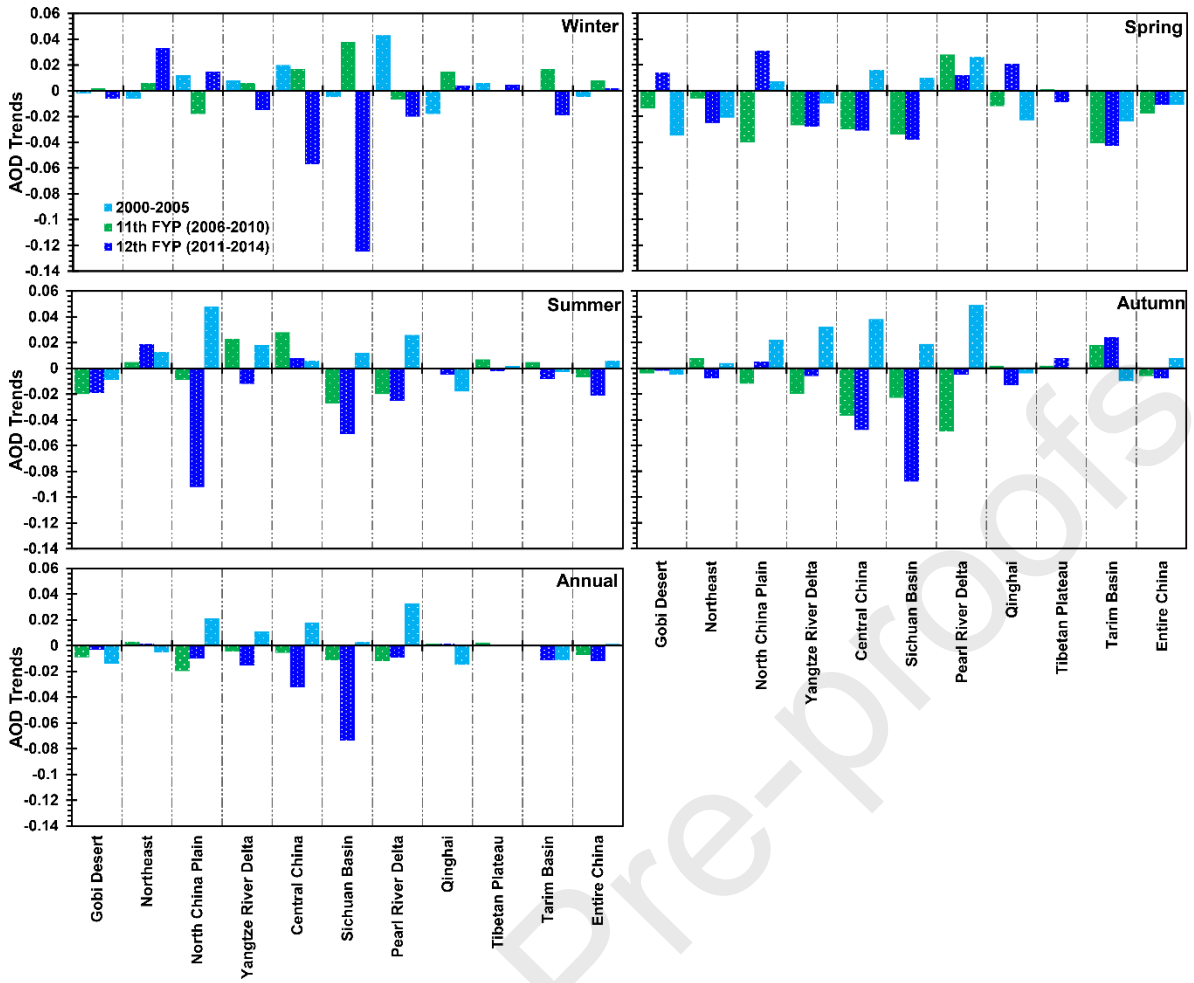
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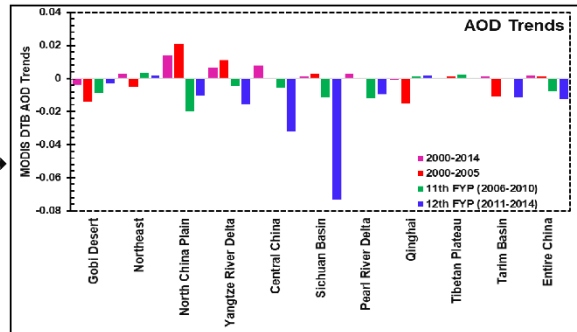
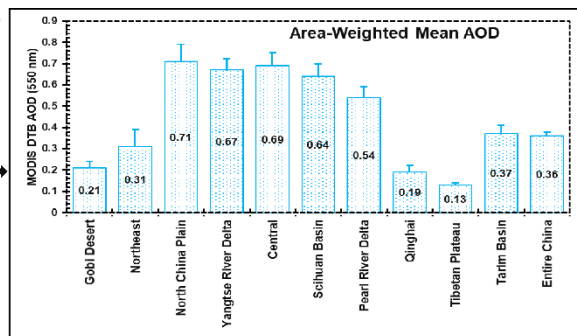
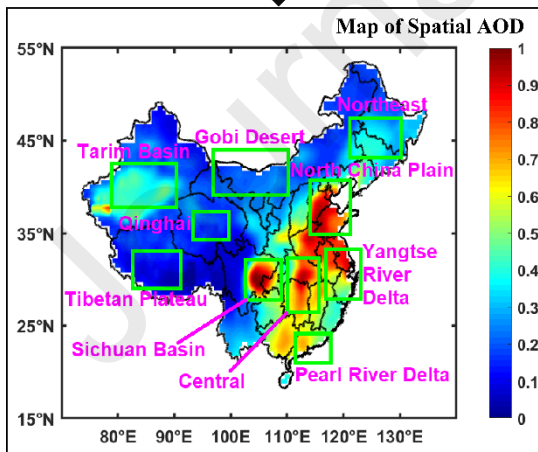


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Temporal changes in aerosol loadings over China from 2000 to 2014



1224

1225 **Highlights:**

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- 1227 • The CMIP6 and MERRA-2 were not in line with Terra-MODIS DTB
- 1228 • AOD upward trends were found during 2000–2014 and 2000–2005
- 1229 • China’s strict air pollution control policies had co-benefits to reduce AOD
- 1230 • AOD reduction was more prominent during the 12th FYP than the 11th FYP period
- 1231 • Seasonally, AOD decrease was more prominent in summer during the 12th FYP
- 1232 period
- 1233

Journal Pre-proofs