

1 **Improvement of the Real-time PM_{2.5} forecast over the**
2 **Beijing-Tianjin-Hebei Region using an optimal interpolation data**
3 **assimilation method**

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12
13 **Abstract**

14 A routine air quality data assimilation (DA) system was established in the China
15 National Environmental Monitoring Center (CNEMC) based on the optimal
16 interpolation (OI) method. The surface observations from more than 1,400 stations
17 over China were assimilated into a real-time air quality forecast system with three
18 nested domains. The initial conditions of NO₂, SO₂ and PM_{2.5} in the three domains
19 were optimized by the data assimilation system. The impact of the data assimilation
20 on the real-time PM_{2.5} forecast over the Beijing-Tianjin-Hebei (BTH) Region during
21 the heavy haze season of 2015 was evaluated. The results show that the DA can
22 significantly improve real-time PM_{2.5} forecasts with the root mean square error
23 (RMSE) reduced by 23%, 8.2%, 4.8% for the forecasts of the first day, second day
24 and the third day respectively. The mean fractional bias and the mean fractional error
25 of the forecast were reduced from 50.9% and 70.67% to 40% and 62.3% respectively,
26 and the performance was changed from "criteria" to approach "goal" (defined by
27 Boylan and Russell, 2006). It is also found that increasing the assimilation frequency

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28 can improve the DA system performance for real-time forecasts. As can be seen from
29 the various case studied here, the improvement of data assimilation is more significant
30 when the bias of the model is higher, and there is still a lot of room for correction. The
31 results also show a rapid decay of the DA effects on the $PM_{2.5}$ forecast, which
32 highlights the limitations of the current routine data assimilation system in which only
33 initial conditions are optimized. Further improvements of the data assimilation system
34 with meteorological data assimilation and chemical parameter optimization are
35 needed.

36 **Keywords**

37 real-time $PM_{2.5}$ forecast, data assimilation, optimal interpolation,
38 Beijing-Tianjin-Hebei Region

39 **1. Introduction**

40 China faces serious atmospheric pollution problems, with high concentrations of
41 fine particulate matter ($PM_{2.5}$) regularly causing serious and large scale haze pollution
42 events (Huang et al., 2014, Li et al., 2016, Wang et al., 2016 Sun et al., 2016). In
43 response to these large-scale haze pollution incidents, in the years since 2013 the
44 China National Environmental Monitoring Center (CNEMC) and many provincial
45 level environmental monitoring departments in China established air quality warning
46 and forecasting system based on the chemistry transport model (CTM). CTM can
47 predict the temporal and spatial distribution of pollutants. But due to the complexity
48 of atmospheric pollution caused by many different pollutants, the uncertainty of
49 emissions and the chemical process modeled in the CTM, air quality forecasts still
50 deviate from actual conditions (Carmichael et al., 2008), in especially heavy pollution
51 the deviation can reach up to 30-50% (Zheng et al., 2015), and the simulation
52 uncertainty of high chemical activity species such as nitrate can reach up to 3 times
53 that (Hayami et al., 2008). DA can reduce the uncertainty (such as its initial
54 conditions) by coupling the model with observations to improve the model
55 performance. (Houtekamer et al., 2005, Tang et al., 2011, Bocquet et al., 2015).

56 DA has thus proven to be an effective method to improve weather forecasts
57 (Bouttier and Courtier, 2002, Kalnay, 2003, Crawford et al., 2016). In the field of air
58 pollution forecasting, Sandu et al. (2011) have shown that data assimilation also plays
59 an important role in improving air pollution prediction. Based on the NAQPMS
60 (Nested Air Quality Prediction Model System, Wang et al., 2001, 2002, 2006), Huang
61 et al. (2016) reanalyzed the $PM_{2.5}$ pollution process in the Beijing-Tianjin-Hebei
62 (BTH) region by using the optimal interpolation assimilation method and found that
63 assimilation can reproduce the pollution process better. Wu et al. (2015) used the
64 3D-Var method to assimilate $PM_{2.5}$ initial conditions base on WRF-Chem (Grell et al.,
65 2005), and improved a 0-48h $PM_{2.5}$ forecast, while Zhen et al. (2017) assimilated the
66 $PM_{2.5}$ initial conditions and emission sources, which can also effectively improve the
67 $PM_{2.5}$ forecast in China base on the WRF-Chem model and ensemble Kalman filter
68 (EnKF) (Evensen, 2010; Schutgens et al., 2010; Yumimoto et al., 2016) EnKF
69 method.

70 In recent years, countries in Europe and other regions have also established
71 assimilation business systems for air quality forecasting (Kukkonen et al., 2012,
72 Marécal et al., 2015). In contrast, in China only air quality prediction systems without
73 DA business systems have been built. Meanwhile, although a large number of air
74 pollutant monitoring sites have been established, their huge data sets are mostly used
75 for *post-hoc* case studies, and have not been used for real-time forecasting. Based on
76 the NAQPMS (Nested Air Quality Prediction Model System, Wang et al., 2001, 2002,
77 2006) and the observation data of 1,436 sites provided by CNEMC, we used the
78 optimal interpolation (OI) method to build a data assimilation business system
79 named ChemDAS for the $PM_{2.5}$ real-time forecast in CNEMC. It began operating in
80 May 2015. In this paper, we added two sets of comparison experiments based on the
81 results of ChemDAS, one is a 72h forecast without DA and the other one is a 72h
82 forecast with different DA frequency, to evaluate the effect of DA for $PM_{2.5}$ real-time
83 forecast (0-72h) during heavy pollution events. The reduction in improvement by DA

84 and the influence of assimilation frequency on real-time forecast are also discussed.

85 **2. Methodology**

86 **2.1 Chemical transport model**

87 In this paper, the chemistry transport model NAQPMS was used, developed by
88 the Institute of Atmospheric Physics, Chinese Academy of Sciences. Based on the
89 three-dimensional Euler sulfide transport model, this model cases emission, advection,
90 diffusion, dry and wet deposition, chemical (including gas phase, aqueous phase,
91 aerosol, and heterogeneous phase) reaction processes, and the incorporation of
92 pollution source tracking, process analysis, and other advanced model techniques, and
93 can carry out multi-scale and multi-pollutant simulation in a mid-latitude area study.
94 NAQPMS settings are consistent with those described in Chen et al. (2015): The
95 improved RADM2 and ISORROPIA1.7 mechanisms are used in the gas phase and
96 inorganic aerosol chemistries (SO_4 , NO_3 , and NH_4). The dust and sea salt processes
97 developed by Luo et al., (2006) and Athanasopoulou et al., (2008) are used to model
98 their respective natural aerosols. The formation of secondary organic aerosols is based
99 on Odum et al., (1997). The heterogeneous chemistry of the aerosol surface is also
100 considered, including 28 chemical reactions (Li et al., 2012). As one of the official
101 CTMs used in CNEMC's air quality prediction system, NAQPMS was successfully
102 used during Beijing's Olympic Games, the Shanghai World Expo, APEC and other
103 important activities in China to ensure air quality (Sun et al., 2016).

104 **2.2 Observation network**

105 Some 1,436 national control observations sites data for $\text{PM}_{2.5}$, SO_2 , NO_2
106 concentrations provided by CNEMC (<http://www.cnemc.cn/>) are used in this paper,
107 and 80 observations sites in the BTH region. As all of these national control stations'
108 observation data are collected within a 1 hour process of automatic collection and
109 upload, it is difficult to avoid errors in the data that may come from individual site
110 abnormalities. On the other hand, as most of the observations sites are concentrated in
111 the eastern part of China, and are densely distributed in urban areas, error data from

112 individual abnormal sites may be diluted by other nearby sites, so it is necessary to
113 pay special attention to the possibility of a larger error skewing the data. To prevent
114 this, we created an automatic data quality control to remove data exceeding the
115 maximum range according to the pollutant measurement instruments ($PM_{2.5} < 1000$
116 $\mu g m^{-3}$, $SO_2 < 1428 \mu g m^{-3}$, $NO_2 < 1026 \mu g m^{-3}$). Fig. 1b shows the distribution
117 of observations sites in D1. For the first time, all country control sites in China are
118 included in one real-time forecasting assimilation system.

119 **2.3 Data assimilation method**

120 The OI method is one of the commonly used assimilation algorithms used to
121 improve the initial conditions of CTMs. Collins et al. (2001) first developed OI to
122 study the inversion of the aerosol optical depth. Wang and Niu (2013) used the OI
123 method to study dust aerosol assimilation in eastern Asian in a mesoscale numerical
124 weather prediction system (GRAPES/CUACE_Dust). A DA system was developed by
125 Jiang et al. (2013) in the WRF-Chem model that used the OI method to study the
126 improvement of DA for PM_{10} simulation over China. Wang et al. (2014) also used the
127 OI method to study the impact of assimilation of lidar observations for aerosol
128 forecasting in the western Mediterranean basin. It was proved that the OI method has
129 lower computational cost than other assimilation algorithms (Wu et al., 2008), which
130 means that it is easier to realize, especially in real-time forecast business systems.
131 4-dimensional variational (4D-Var) and the ensemble Kalman filter (EnKF) are two
132 other widely used assimilation methods with better assimilation effects than OI
133 method (Benedetti and Fisher, 2007, Candiani et al., 2013), but 4D-Var requires a
134 more complex concomitant model (Benedetti et al., 2009, Sugimoto and Uno, 2009),
135 while the computational cost of EnKF is much larger than an OI method (Denby et al.,
136 2008, Pagowski and Grell 2012). Taking into account the timeliness and
137 computational cost in real-time forecasting, our system used the OI method.

138 The OI approach uses the optimal linear combination between the background
139 state and the observed value (Daley, 1991). The analytical values can be obtained

140 from the following equation

$$141 \quad x_a = x_b + \mathbf{BH}^T(\mathbf{HBH}^T + \mathbf{R})^{-1}(y - H[x_b]) \quad (1)$$

142 In this equation, x_a is the analyzed mass concentration, x_b is the background
143 vector (model mass concentration), y is the observation vector, H is the observation
144 operator, \mathbf{H} is the tangent linear operator of the observation operator H , \mathbf{B} is the
145 background error covariance matrix with static assumptions, and \mathbf{R} is the observation
146 error covariance matrix. Only a few observation data are important for incremental
147 decision analysis in generally, which means it only assimilates patterns of observation
148 information around a model coordinate point. For the selection of the observation
149 error covariance matrix \mathbf{R} , the observation error variance is $\delta^2 = [0.1Y(i)]^2$, and the
150 observation error mean variance is 15% for all the observation points. In addition, the
151 spatial correlation of the observation site is ignored, so \mathbf{R} is a diagonal matrix. This
152 article used a static and isotropic background error covariance, and the error
153 correlation between state variables of each grid only depends on the space distance
154 between each grid. The horizontal correlation function uses a Gaussian distribution
155 function:

$$156 \quad Cov_{i,j}(d) = \left(1 + \frac{d}{L}\right) e^{-\frac{d}{L}} Var_i \quad (2)$$

157 L is the characteristic scale of the spatial correlation. According to the model
158 resolution and spatial distribution of the observation site, 125 km was selected for this
159 paper. d is the distance between two grids, and Cov is the error covariance between
160 two grids. Var is the model simulation error variance between two grids, according to
161 a long term comparison between the simulation and observation, it was set to 80% of
162 the benchmark simulation concentration.

163

164 **2.4 Configuration of the routine PM_{2.5} data assimilation**

165 In this system, the NAQPMS model uses a three-layer nested domain. As shown
166 in Fig. 1a, the first domain (D1) covers the entire East Asian region, and the

167 horizontal resolution is 45 KM, the second domain (D2) covers much of China's
168 landmass, and the horizontal resolution is 15 KM. Hebei province is the center of the
169 third domain (D3) which includes the BTH region, Henan, Shanxi, Shandong,
170 Liaoning and other neighboring provinces, the horizontal resolution of D3 is 5 KM.
171 The model uses a Sigma-Z terrain-following coordinate, the vertical layer of
172 1000-100hPa was divided into 20 layers with 8 layers under 2 KM. In this study, we
173 mainly evaluate the improvement of simulation in the BTH region, so all the results
174 presented in this paper are from D3.

175 Multi-Resolution Emission Inventory for China (MEIC, <http://www.meicmodel.org/>
176) developed by Tsinghua University was used for anthropogenic sources. The
177 resolution is $0.25^{\circ} \times 0.25^{\circ}$ and the base year is 2010. Biogenic emissions were taken
178 from the Global Emission Inventory Activity (GEIA) (Guenther et al., 1995) and
179 biomass burning emissions were from Cao et al., (2005).

180 The mesoscale meteorological model WRFv3.6 was used for calculating the
181 hourly model meteorological field, and the meteorological initial and boundary
182 conditions were obtained from the National Centers for Environmental Prediction
183 Global Forecast System (GFS). In this paper, the experiment period was November
184 1-December 3, 2015 during which there were three cases of heavy pollution over the
185 BTH region. At every 0 o'clock (UTC) and 12 o'clock (UTC) a prediction was made
186 using the previous prediction as a baseline. The model integration time step was 5
187 min and the output frequency was 1 h.

188 All the initial conditions of the three domains were assimilated during the
189 simulation period. In the vertical direction, the range of assimilation was 3 layers. As
190 SO_2 and NO_2 are related to the $\text{PM}_{2.5}$ precursor, we assimilated $\text{PM}_{2.5}$, SO_2 and NO_2
191 in the initial field. In the initial field, the variables related to $\text{PM}_{2.5}$ concentration were
192 $\text{PM}_{2.5}$, BC, OC, SOA1, SOA2, SOA3, SOA5, SOA6, NH_4AQ , SO_4AQ , HSO_4AQ ,
193 NO_3AQ , $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , $\text{H}_2\text{SO}_4\text{AQ}$, $\text{NH}_4\text{HSO}_4\text{S}$, and $(\text{NH}_4)_4(\text{HSO}_4)_2$, a total of
194 18 components in NAQPMS. We first distributed observed $\text{PM}_{2.5}$ concentrations to all

195 the related components, according to the 18 components concentration ratio in the
 196 original initial field, then assimilated all the components respectively.

197 In order to evaluate the effect of assimilation for real-time forecasting, we added
 198 two sets of simulation experiments shown in **Table 1**. Control (CT) was a 72h
 199 forecast without DA at every 12 o'clock (UTC). 24h-DA provided 72h forecasting
 200 with DA at every 12 o'clock (UTC) to evaluate the impact of assimilation frequency
 201 for 72h forecasting, and 12h-DA was the same as the system setup. All of the
 202 experiment's first forecasts were based on the same initial conditions before DA,
 203 which was taken from the business system.

204 **Table 1** Experiments setup

Experiment	Forecasting setup	DA method	DA time
Control (Model without DA)	72 hours at 12 o'clock (UTC)	-	-
24h-DA	72 hours at 12 o'clock (UTC)	OI	12 o'clock (UTC)
12h-DA	72 hours at 12 o'clock (UTC), 12 hours at 0 o'clock (UTC)	OI	12 o'clock (UTC), 0 o'clock (UTC)

205

206 **3. Results and discussion**

207 **3.1 PM_{2.5} pollution episodes**

208 From November 1st to December 3rd, 2015, most regions in the BTH region were
 209 heavily polluted as a whole (PM_{2.5} average concentration up to 95 $\mu\text{g m}^{-3}$). Beijing,
 210 Baoding, Langfang and some other cities' PM_{2.5} daily average concentration reached
 211 500 $\mu\text{g m}^{-3}$ or more. **Fig. 2** shows the time series of daily average PM_{2.5}
 212 concentration of all cities in the BTH region during the simulation period. The red
 213 line represents the average in the BTH region and the shadowed areas represent cities
 214 Beijing, Tianjin, Shijiazhuang and all the other cities in Hebei Province. As can be
 215 seen in **Fig. 2**, the different cities experienced different degrees of haze pollution
 216 during the simulation period, but the aggregate trend is consistent. In addition to
 217 assessing the effect of assimilation over the whole period, we also wanted to know the

218 effect of assimilation during each process of the pollution event, so we divided the
219 whole simulation time into three cases according to changes in $PM_{2.5}$ concentration.
220 Among them, Case 2 was the longest-running case, lasting 10 days in total, and Case
221 3 was the most serious case, with the daily average concentration of $PM_{2.5}$ reaching
222 up to $250 \mu g m^{-3}$ or more in the whole BTH area.

223

224 3.2 Improvement of initial conditions

225 We chose D3 for the initial conditions verification because at the highest
226 resolution (5 KM) it allows the observed data to distribute as much as possible in the
227 different grids. Therefore, all the observations sites (490) in the D3 were divided into
228 two groups averagely in each city, one for assimilation and the other group for
229 verification. These sites are shown in **Fig. 3a** (black point sites are those used for
230 assimilation and red circle sites are those used for verification). **Fig. 3b** shows the
231 scatter plot comparison of $PM_{2.5}$ concentrations of the simulation and actual
232 observations both before and after assimilation in D3 on October 31st. It is obvious
233 that the assimilation corrected the initial $PM_{2.5}$ concentration significantly, especially
234 in the region overestimate, and the RMSE of all verification sites was reduced from
235 $100 \mu g m^{-3}$ to $53.2 \mu g m^{-3}$. Comparison of the RMSE of every 72h forecast's
236 initial conditions during experimental period before and after DA (12h-DA
237 experiment, all the same in Section 3.2, **Fig. 3** and **Fig. 4**) is shown in **Fig. 3c**. The
238 improvement of initial conditions with DA was significant for the majority of these
239 verifications, except on November 8th. The reason for the high value of RMSE before
240 and after DA on November 8th might be that the observed data include a sudden
241 increase in most of Tianjin's sites, further illustrating the importance of the
242 observations' data quality for DA. In addition, as **Fig. 4** shows, we compared the
243 $PM_{2.5}$ observations and $PM_{2.5}$ concentration in initial conditions before and after DA
244 within all the 1,436 observations sites in every domain on October 31st. This was done
245 to understand the impact of DA for initial conditions in the system's actual operating
246 situation. In D1 and D2, the improvement of the simulation in the southwest region of

247 China is obvious, while in D3 the distribution of $PM_{2.5}$ concentration is closer to the
248 observations distribution after DA.

249

250 3.3 Improvement of the 24h $PM_{2.5}$ forecast

251 **Fig. 5** shows the time series of the 24h real-time forecast $PM_{2.5}$ daily average
252 concentrations of the three experiments in Beijing during experimentation period, with
253 the statistical related coefficient (R) and RMSE, in which a represents CT, b represents
254 24h-DA, and c represents 12h-DA. It can be seen from the figure that the correlation
255 coefficients of the three experiments were both satisfactory ($R > 0.82$) in Case 1, and
256 the RMSE of 12h-DA was reduced from 61.3 (CT) to 45.9, in Case 2, the relative
257 coefficient of 24h-DA and 12h-DA had a small improvement compared to CT, while
258 the RMSE both improved remarkably (12h-DA's RMSE decreased by 37.4%).
259 However, the RMSE after assimilation was still high ($RMSE > 100$), which means the
260 assimilation effect still had a larger optimized space. Case 3 had a smaller
261 assimilation effect compared with the other two cases. In general, the assimilation
262 improvement effect of the 24h real-time forecast was acceptable for the heavy
263 pollution event in Beijing, especially for the RMSE, and the improvement effect is
264 more obvious when the simulation deviation is larger. In addition to the northern
265 cities such as Zhangjiakou and Chengde, most of cities in the BTH region had similar
266 characteristics with Beijing. Besides this, the assimilation improvement effect of
267 12h-DA was also better than 24h-DA. **Fig. 6** shows the distribution of $PM_{2.5}$ monthly
268 mean concentrations of observations and three experiments in D3 during November
269 2015. The simulated concentration of $PM_{2.5}$ in the whole BTH region is significantly
270 reduced after assimilation, which is in good agreement with the spatial distribution of
271 the $PM_{2.5}$ observations. Here the improvement effect by 12h-DA is also better than
272 24h-DA.

273

274 According to the study of Boylan and Russell (2006), the mean fractional bias

275 (MFB) and the mean fractional error (MFE) can be used to judge model performance.
 276 When ~~Both~~ both MFE and MFB are less than or equal to +75% and $\pm 60\%$
 277 respectively, the model performance “criteria” has been met. Additionally, the model
 278 performance “goal” has been met when both the $MFE \leq +50\%$ and $MFB \leq \pm 30\%$.
 279 **Table 2** shows the statistics of the 24h PM_{2.5} real-time forecast results of three
 280 experiments in the BTH region that lasted from November 1st to December 3rd 2015.
 281 From the average of the observations (MO) and the average of the model results
 282 (MM), it is clear that the model estimation was higher than the observations over the
 283 whole the BTH region, and the model bias (MB) was big. It also can be seen from the
 284 table, the model performance is “criteria” level without DA (MFB is 50.9% and MFE
 285 is 70.7%). The model performance was improved in the 24h-DA experiment: the
 286 MFB and MFE were both reduced and the RMSE was reduced by about 16% in BTH.
 287 Compared to the 24h-DA experiment, the model performance of the 12h-DA
 288 experiment was further improved, the RMSE was reduced by about 10%, and the
 289 MFB and MFE are further approximated to the "goal" level, so increasing
 290 assimilation frequency can help to improve the assimilation effect. The results for
 291 Beijing, Tianjin and Shijiazhuang also show with the same characteristics. Among
 292 them, the model performance in Tianjin is closest to the "goal" level with DA, and the
 293 assimilation helped the model performance in Shijiazhuang to reach the “criteria”
 294 level.

295 **Table 2.** The Statistic of 24h PM_{2.5} real-time forecast results of three experiments in BTH from
 296 November 1st to December 3rd

	Experiment	MO ($\mu\text{g m}^{-3}$)	MM ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	RMSE	MFB	MFE
BTH	Control	95.0	156.7	61.7	101.4	50.90%	70.67%
	24h-DA	95.0	142.0	47.0	85.0	44.90%	65.39%
	12h-DA	95.0	132.2	37.1	77.5	40.00%	62.30%
Beijing	Control	122.2	187.5	65.3	101.1	51.30%	68.36%

	24h-DA	122.2	170.1	47.9	82.7	44.50%	62.01%
	12h-DA	122.2	160.9	38.7	76.3	40.00%	59.37%
Tianjing	Control	90.3	148.9	58.6	81.2	50.70%	64.22%
	24h-DA	90.3	137.1	46.9	69.4	44.80%	58.94%
	12h-DA	90.3	128.6	38.3	62.9	40.00%	55.97%
Shijiazhuang	Control	111.2	214.2	102.9	123.8	72.10%	80.19%
	24h-DA	111.2	185.7	74.4	97.8	62.90%	72.05%
	12h-DA	111.2	170.0	58.8	86.3	57.00%	67.98%

297

298 3.4 Improvement of the 72h PM_{2.5} forecast

299 The simulation effect of 72h real-time forecast is important for air quality
300 prediction, so it is necessary to evaluate the assimilation improvement effect of the
301 72h real-time forecast. **Fig. 7** shows the 0-72 forecast hour PM_{2.5} concentrations of
302 observations, a CT and 12h-DA experiment, and the RMSE of CT and a 12h-DA
303 experiment in both a single real-time forecast (on November 5th) and over the whole
304 experiment period (with averages corresponding to the same forecast hour) in BTH.
305 In both the single real-time forecast and the whole experiment period, the assimilation
306 effect on the 0-24h forecast was significantly higher than that of 24-48h forecast, the
307 assimilation improvement effect on the 48-72h forecast was even weaker. The
308 assimilation had an impact on all of the forecasted hours, but the improvement effect
309 was diminished as time increased. To further study the assimilation improvement
310 effect of every forecast hour in a 72h real-time forecast, we compared the attenuation
311 curve of the RMSE improvement effect of 12h-DA experiment with CT in BTH (as
312 showed in **Fig. 8**). The RMSE improvement is best (20%-65%) in the initial stage of
313 forecast (<12h), but the improvement of RMSE rapidly fell to about 15% in the 24th
314 forecast hour, then further dropped to about 10% during the 24-48h period, and finally
315 became weak (<10%) during the 48-72h period.

316 **Table 3** shows the statistics of the RMSE improvement of the 12-DA experiment

317 compared with the CT for 72h real-time forecast in the BTH region. For the 0-24h
 318 real-time forecast, the improvement effect of RMSE was 23.6% in the whole of the
 319 BTH region, and higher than 20% in most cities. Handan had the highest rate (33.7%),
 320 and Zhangjiakou the lowest (16.8%) because of the lowest background concentration
 321 of PM_{2.5}. For the 24-48h and 48-72h real-time forecasts, the RMSE improvement was
 322 8.2% and 4.8% in BTH respectively, with about 10% and 5% in cities. For the whole
 323 72h real-time forecast, the RMSE improvement was 12.8% in the BTH region and
 324 about 10% in cities.

325 **Table 3** the statistics of the RMSE improvement of 12-DA experiment compared with CT for 72h
 326 real-time forecast in BTH

	0-24h (%)	24-48h (%)	48-72h (%)	0-72h (%)
JJJ	23.6	8.2	4.8	12.3
Beijing	24.5	6.2	5.3	12.1
Tianjing	22.5	8.6	7.1	12.8
Shijiazhuang	30.3	10.3	7.0	16.2
Tangshan	16.2	4.8	5.7	9.2
Qinhuangdao	17.2	8.4	6.6	10.9
Handan	33.7	12.4	5.2	17.8
Xingtai	32.6	10.2	5.9	16.8
Baoding	22.9	7.8	0.7	10.6
Zhangjiakou	16.8	9.1	6.8	10.9
Chengde	18.4	9.2	5.6	11.0
Cangzhou	21.0	11.4	9.0	13.8
Langfang	17.1	5.8	2.5	8.7
Hengshui	22.3	10.9	3.7	12.3

327

328 4. Conclusions

329 In this paper, we built a data assimilation business system for 72h real-time

330 forecasts with CNEMC's 1,436 national control observation sites data and an OI
331 method. This was the first such system in China. The initial conditions of PM_{2.5}, SO₂,
332 NO₂ concentrations are both assimilated into this system. To evaluate the
333 improvement of the DA system for PM_{2.5} simulation, three experiments during PM_{2.5}
334 heavy pollution in the BTH region were set. We found that the model bias of 24h
335 real-time forecast in the BTH region was higher ($61.7 \mu\text{g m}^{-3}$) without DA and
336 lower ($37.1 \mu\text{g m}^{-3}$) with DA, meanwhile the RMSE was reduced from 101.4 before
337 assimilation to 77.5 afterwards. The model performance had been approach "goal"
338 from "criteria" with the MFB and MFE was reduced from 50.9% and 70.67% to 40%
339 and 62.3% respectively. Comparing this to a 24h-DA experiment (which assimilated
340 once a day), the 12h-DA (which assimilated twice a day) showed greater
341 improvement (about 10% for RMSE), which means that increasing the assimilation
342 frequency can improve the DA system performance for real-time forecasts. The
343 RMSE improvement is 24%, 8.2%, 4.8% and 12.3% for 0-24h, 24-48h, 48-72h and
344 0-72h in BTH, respectively. The improvement effect was diminished as the forecast
345 hour increased, especially in the beginning (0-12h). The RMSE improvement in first
346 24h forecast stayed above 15%, then dropped to about 10% during the 24-48h
347 forecast, and finally became weak (<10%) during the 48-72h period.

348 The RMSE was over 100 with DA during CASE 2. Even this RMSE
349 improvement is significant, possibly because of the uncertainty of the emission
350 sources and the physical and chemical mechanisms in the model, but the fact that the
351 RMSE of initial conditions was still high after assimilation is likely one of the reasons
352 for this result. This means that even if the assimilation improvement effect is
353 acceptable as a whole, the system has a larger optimized space. It is necessary to
354 improve the DA system in terms of the parameters of the OI algorithm, assimilation
355 frequency, observation data quality control, and so on, in the future.

356 **Acknowledgements**

357 This work was supported by the National Natural Science Foundation [Grant No.

358 41575128, 91544218, 91644216].

359

360 **References**

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