1 **Improving the Sectional MOSAIC Aerosol models of WRF-Chem with the**
2 **revised Gridpoint Statistical Interpolation System and multi-wavelength aerosol**
3 **optical measurements: DAO-K experiment 2019 at Kashi, near the Taklamakan**
4 **Desert, northwestern China**
5
6 Wenyuan Chang[1], Ying Zhang[2], Zhengqiang Li[2], Jie Chen[3], Kaitao Li[2]
7
8
9 [1] State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry
10 (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029,
11 China.
12
13 [2] Institute of Remote Sensing and Digital Earth, Chinese Academy of Sciences, Beijing
14 100101, China
15
16 [3] National Meteorological Information Center, China Meteorological Administration, Beijing
17 100081, China
18
19
20
21
22
23 Corresponding authors:
24 Wenyuan Chang (changwy@mail.iap.ac.cn)
25 Zhengqiang Li (lizq@radi.ac.cn)
26
27

28   **Abstract**

29   The Gridpoint Statistical Interpolation data assimilation (DA) system was developed for the

30   four-size bin sectional Model for Simulating Aerosol Interactions and Chemistry (MOSAIC)

31   aerosol mechanism in the Weather Research and Forecasting-Chemistry (WRF-Chem) model.

32   The forward and adjoint operators for the aerosol optical depth (AOD) analysis were derived

33   from WRF-Chem aerosol optical code. We applied three-dimensional variational DA to

34   assimilate the multi-wavelength AOD, ambient aerosol scattering coefficient, and aerosol

35   absorption coefficient, measured by the sun-sky photometer, nephelometer, and aethalometer,

36   respectively. These were undertaken during a dust observation field campaign at Kashi in

37   northwestern China in April 2019. The results showed that the DA analyses decreased the low

38   biases in the model aerosols; however, it had had some deficiencies. Assimilating the surface

39   particle concentration increased the coarse particles in the dust episodes, but AOD, and the

40   coefficients for aerosol scattering and absorption, were still lower than observed values.

41   Assimilating aerosol scattering coefficient separately from AOD improved the two optical

42   quantities. However, it caused an overestimation of the particle concentrations at the surface.

43   Assimilating the aerosol absorption coefficient yielded the highest positive bias in the surface

44   particle concentration, aerosol scattering coefficient, and AOD. The positive biases in the DA

45   analysis were caused by the forward operator underestimating particle scattering and

46   absorption efficiency. As a compensation, the DA system increased particle concentrations

47   excessively so as to fit the observed optical values. The best overall improvements were

48   obtained from the simultaneous assimilation of the surface particle concentration and AOD.

49   The assimilation did not substantially change the aerosol chemical fractions. After DA, the

50   clear-sky aerosol radiative forcing at Kashi was -10.5 Wm^{-2} at the top of the atmosphere,

51   which was 46% higher than the background radiative forcing value.

1. Introduction

Data assimilation (DA) blends the information from observations with *a priori* background
fields from deterministic models to obtain an optimal analysis (Wang et al., 2001; Bannister,
2017). With lagged emission inventories and unsatisfactory model chemistry mechanisms,
there are notable discrepancies between model aerosols and observed levels (He et al., 2017;
Chen L. et al., 2019). The DA technology incorporates aerosol measurements into the models
to optimize emissions (Peng et al., 2017; Ma et al., 2019), and cyclically updates the
background fields in forecasts. This effectively improves the air quality forecasts in China
(Bao et al., 2019; Cheng et al., 2019; Feng et al., 2018; Hong et al., 2020; Liu et al., 2011;
Pang et al., 2018; Peng et al., 2018; Xia et al., 2019a, 2019b).

Variational DA minimizes the distant scalar function measuring the misfit between model
states and a set of observations in each assimilation window. An effective variational DA
requires an appropriate adjoint operator (or Jacobian matrix), which describes the gradient or
sensitivity of the observed parameter to the control variable (Wang et al., 2001; Bannister
2017). The adjoint operator is highly dependent on the types of assimilated observations and
the selection of control variables; it is also sometimes dependent on the aerosol mechanism.
For $PM_{2.5}$ (particulate matter with dynamic radius less than 2.5 µm) DA, the adjoint operator
is the ratio of the $PM_{2.5}$ concentration to composition of each aerosol (Pagowski et al., 2010).
For the aerosol optical depth (AOD) DA, the adjoint operator is generated through Mie theory
(Liu et al., 2011; Saide et al., 2013). With the development of aerosol mechanisms and the
growing body of novel aerosol observations from ground-based networks and satellites, an
appropriate adjoint operator is in demand.

The community gridpoint statistical interpolation (GSI) system (Wu et al., 2002; Purser et al.,
2003a, 2003b) is often used to modify regional aerosol simulations with three-dimensional
variational (3D-Var) DA. The official GSI (version 3.7 in this study) can incorporate
observations of surface particulate matter concentration and AOD to constrain the aerosols
simulated within the aerosol mechanism of Goddard Chemistry Aerosol Radiation and
Transport (GOCART, Liu et al., 2011; Pagowski et al., 2014). The tangent linear operator and
adjoint operator for AOD were determined using the Community Radiative Transfer Model
(CRTM). This GSI version incorporating the Moderate Resolution Imaging
Spectroradiometer (MODIS) AOD in East Asia (Liu et al., 2011), revealed the simultaneous
DA effects of $PM_{2.5}$ and AOD in the continental United States (Schwartz et al., 2012). This
GIS was used to identify DA effects that weakened during running of the succeeding model
as the model error grew (Jiang et al., 2013), and assessed the radiative forcing of the aerosols
released by wildfires (Chen et al., 2014). This version of GSI was also utilized to improve air
quality forecasts in China by assimilating a variety of satellite AOD data retrieved from: the
Geostationary Ocean Color Imager (Pan et al., 2018); Visible Infrared Imaging Radiometer
Suite (Pang et al., 2018); Advanced Himawari-8 Imager (Xia et al., 2019a); and the Fengyun-
3A/medium-resolution spectral imager (Bao et al., 2019; Xia et al., 2019b).

Despite its capabilities, the GOCART mechanism is unable to simulate nitrate and secondary
organic aerosols (SOA), and the GOCART aerosol size distribution uses a bulk assumption

96   for radiative transfer calculation. Strictly speaking, the lack of aerosol components violates
97   the unbiased requirements for the model states in the DA system. Lack of size-segregated
98   aerosols may introduce a bias in the calculation of optical aerosols. The official GSI can
99   assimilate the surface particle concentration from the aerosol mechanism apart from
100 GOCART (Zang et al., 2016), but its AOD DA is tightly bound with the GOCART aerosols.
101 If one wished to use GSI to assimilate AOD for the other aerosol mechanisms, a compromise
102 solution was to integrate the map of the speciated aerosols of other mechanisms into that of
103 the GOCART aerosols. For example, Tang et al. (2017) used the official GSI to assimilate
104 MODIS AOD with the aerosols from the Community Multi-scale Air Quality Model
105 (CMAQ). They incorporated the map of the 54 aerosol components of CMAQ into the five
106 CRTM aerosols and repartitioned the mass increments of each CMAQ aerosol according to
107 the ratio of aerosol chemical components in the background field. This repartitioning is called
108 the "ratio approach." Cheng et al. (2019) assimilated the lidar extinction coefficient profiles
109 measured in Beijing to modify the Weather Research and Forecasting-Chemistry (WRF-
110 Chem) Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosols. They
111 used the ratio approach to map eight MOSAIC aerosols based on five GOCART aerosols.
112 This mapping strategy is readily implemented but introduces inconsistent size-segregated
113 aerosol information (e.g., hygroscopicity and extinction efficiency) between the aerosol
114 model and the DA system. Because building a GSI system for a new aerosol mechanism is
115 quite technical, the official GSI for the GOCART aerosols is still a primary choice for recent
116 aerosol DA studies (Bao et al., 2019; Xia et al., 2019; Hong et al., 2020).

118 Because of the shortcomings, the official GSI has been extended to cooperate with other
119 aerosol mechanisms in WRF-Chem. The MOSAIC mechanism in WRF-Chem simulates
120 aerosol mass and number concentrations in either four- or eight-size bins. This sectional
121 aerosol mechanism involves nitrate chemistry and can simulate SOA with the volatility basis
122 set scheme. Saide et al. (2013) proposed a revised GSI version that performed variational DA
123 for the MOSAIC aerosols. The authors generated the adjoint operator code with the automatic
124 differentiation tool (ADT), TAPENADE v3.6. The ADT used the chain rule of derivative
125 calculus on the AOD source code in WRF-Chem. They assimilated multi-source AOD data
126 with the MOSAIC aerosols over continental United States and found that incorporating multi-
127 wavelength fine-mode AOD redistributed the aerosols' particulate mass concentration sizes.
128 The revised GSI system assimilated Korean ground-based and geostationary satellite AOD
129 datasets to improve local aerosol simulations (Saide et al., 2014, 2020). Pang et al. (2020)
130 developed the official GSI to work with the Modal Aerosol Dynamics Model for Europe with
131 the Secondary Organic Aerosol Model (MADE/SORGAM) aerosols in WRF-Chem. The
132 authors used the WRF-Chem AOD code as the forward operator to calculate the essential
133 aerosol optical properties, which were then inputted to the CRTM adjoint operator. Because
134 aerosols were externally mixed in CRTM, the setting of the internal mixture per size bin in
135 WRF-Chem was not taken into account, and the AOD of each aerosol component was
136 calculated separately.

138 This study provides a solution to improve the capability of the GSI 3D-Var DA system for the
139 sectional MOSAIC aerosols in WRF-Chem. We designed the adjoint operator code for AOD

140   DA based on the WRF-Chem intrinsic aerosol optical subroutine (Fast et al., 2006), that is,

141   without using the ADT. The adjoint code is programmed based on the analytical equations of

142   the linear tangent model for AOD. As our revised GSI does not use the CRTM module, it

143   avoids the problem of needing to eliminate WRF-Chem aerosols characteristics (e.g., aerosol

144   mixture state and size distribution) to meet the CRTM input requirements. The forward and

145   adjoint operators are coordinated, since they are derived from the same WRF-Chem code, and

146   are written in a single subroutine, which is coupled to the GSI at the place of invoking CRTM

147   for the AOD calculation. In addition to AOD DA, our adjoint operator has two variants to

148   assimilate the aerosol scattering and absorption coefficients, measured using a nephelometer

149   and aethalometer, respectively.

150

151   This study verifies the effectiveness of our revised GSI system by incorporating multi-

152   wavelength aerosol optical observations that were measured during an international field

153   campaign, the Dust Aerosol Observation-Kashi, in April 2019 at Kashi city, neighboring the

154   Taklamakan Desert, northwestern China. This desert is the second largest globally, and is the

155   primary source of dust aerosols in East Asia. The dust from the desert affects the nearby

156   Tibetan Plateau (Ge et al., 2014; Jia et al., 2015; Zhao et al., 2020), air quality and climate in

157   East Asia (Huang et al., 2014), and the biogeochemical cycles in the western Pacific Ocean

158   (Calil et al., 2011). A successful DA analysis will help improve the local air quality forecast

159   and enhance our understanding of the environmental impacts of local dust storms. The

160   remainder of this paper is organized as follows. Section 2 describes the revised GSI system,

161   the experimental design, and the observed data. Section 3 presents the DA results when

162   assimilating different observations. Section 4 discusses the impact of DA on aerosol chemical

163   composition and aerosol direct radiative forcing. Finally, Section 5 provides the conclusions

164   and limitations that need further research.

165

166   **2. Methodology and Data**

167   **2.1 Forecast Model**

168   The background aerosol fields were simulated using the WRF-Chem model version 4.0 (Grell

169   et al., 2005; Fast et al., 2006). The model configurations included the Purdue Lin

170   microphysics scheme (Chen and Sun, 2002), the unified Noah land surface model (Tewari et

171   al., 2004), the Yonsei University scheme for planetary boundary layer meteorological

172   conditions (Hong et al., 2006), and the rapid radiative transfer model for general circulation

173   models (RRTMG) scheme for shortwave and longwave radiation (Iacono et al., 2008). The

174   gas-phase chemistry was simulated using the carbon bond mechanism (Zaveri and Peters,

175   1999), including aqueous-phase chemistry. The aerosol chemistry was simulated using the

176   MOSAIC mechanism (Zaveri et al., 2008), which simulated sulfate, nitrate, ammonium, black

177   carbon (BC), organic carbon (OC), sodium, calcium, chloride, carbonate, and other inorganic

178   matter (OIN, e.g., trace metals and silica). SOA was excluded from our experiments to

179   accelerate model integration. Although ignoring that SOA biased the model, the influence was

180   assumed to be small, based on low anthropogenic and biogenic emissions in the vicinity of the

181   desert. We performed the MOSAIC aerosol simulations with four-size bins (0.039–0.156 μm,

182   0.156–0.625 μm, 0.625–2.500 μm, and 2.5–10.0 μm dry diameters). The sectional aerosol

183   data in the hourly model output were the aerosol dry mass mixing ratios of chemical

184 compositions, aerosol number concentration, and aerosol water content. The aerosol
185 compositions included hydrophilic particulates (i.e., SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Na^+) and
186 hydrophobic particulates (i.e., BC, OC, and OIN). The dust emission was simulated using the
187 GOCART dust scheme (Ginoux et al., 2001). The dust mass was included in the OIN
188 concentration determination and aerosol optical calculation. The aerosol compositions were
189 externally mixed between the size bins and internally mixed in each size bin. The internal
190 mixing refractive index was the volume-weighted mean refractive index of each composition.
191

192 **2.2 Assimilation System**
193 The revised GSI DA system is based on the official GSI (https://dtcenter.org/community-
194 code/gridpoint-statistical-interpolation-gsi, Wu et al., 2002; Liu et al., 2011; Schwartz et al.,
195 2012; Pagowski et al., 2014) version 3.7. The 3D-Var DA minimizes the cost function:
196

197 $$J(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_b)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + \frac{1}{2}(H(\mathbf{x}) - \mathbf{y})^T \mathbf{R}^{-1}(H(\mathbf{x}) - \mathbf{y})$$

198 (1)
199

200 where $\mathbf{x}$ is the state vector composed of the model control variables; the subscript b denotes
201 that $\mathbf{x}$ is the background state vector; $\mathbf{y}$ is the vector of the observations; H is the forward
202 operator or observation operator that transfers the gridded control variables into the observed
203 quantities at the observation locations; and $\mathbf{B}$ and $\mathbf{R}$ are the background and observation error
204 covariance matrices, respectively.
205

206 The official GSI version only works with the GOCART aerosols for assimilating the surface-
207 layer $PM_{2.5}$ and PM_{10} (denoted as PM_x in the context) concentrations, and the 550 nm MODIS
208 AOD. Our revised GSI system assimilates PM_x concentrations, multi-wavelength aerosol
209 scattering/absorption coefficients, and AOD. Figure 1 shows the workflow of our DA system.
210 According to the AOD calculation in WRF-Chem, we can either choose the aerosol number
211 concentration (option 1), or aerosol mass concentration (option 2) as control variables. Option
212 1 is described in Li et al. (2020). In this study, we selected option 2, which is described in the
213 following subsections.
214

Figure 1

215

216 **2.2.1 Control Variables**
217 The control variables in this study were the mass mixing ratio of composition of each aerosol
218 per size bin, which corresponded to the WRF-Chem output data only. This set therefore
219 differed from previous studies that lumped aerosols per size bin as control variables. The
220 lumped aerosols avoided the burdensome task of specifying the background error statistics for
221 numerous aerosols (Li et al., 2013; Pagowski et al. 2014). Although our control variables
222 could have been further optimized, here we designed the control variable using only those that
223 substantially contributed to the total mass concentrations. In the case of Kashi situated near
224 the desert, the OIN was predominant, accounting for ~99% of the total particle mass
225 concentrations. The control variable could thus have exclusively comprised the OIN.

Atmospheric
Chemistry
and Physics
Discussions

Open Access

EGU

226 However, because we were curious about the response of aerosol chemical fractions in the
227 DA constraint, we set the control variables of five aerosol mass mixing ratios of SO_4^{2-}, NH_4^+,
228 OC, BC, and OIN per size bin. Nitrate, chlorine, and sodium had miniscule background
229 concentrations and remained the background values in the DA analysis. There were twenty
230 control variables in total for the four-size bin simulations, and the time cost for the DA
231 calculation for these variables was acceptable.
232
233 Our design of the control variables was different from the AOD assimilation in Saide et al.
234 (2013), with theirs being the natural logarithm of the total mass mixing ratio per size bin,
235 multiplied by the thickness of the model layer. As the high model layer had a significant layer
236 thickness with low aerosol concentrations, the multiplication offset the opposite effects of
237 increasing layer thickness versus decreasing concentrations with increase in altitude. This
238 prevented the addition of many modifications for the high model layers, where aerosols were
239 low in concentration. The logarithmic transformation was used to decrease the extensive
240 value range in the control variables caused by multiplication. Since the AOD value is often
241 smaller than one, this leads to a significant negative logarithm value and a relatively
242 unconstrained DA system. Saide et al. (2013) introduced two weak constraints in their cost
243 function to cut off the user-defined "extraordinarily high" and "extraordinarily low"
244 concentrations. However, neither the logarithmic transformation, nor the multiplication using
245 layer thickness was set in our DA system. Saide et al. (2013) repartitioned the increments of
246 the total mass per size bin for composition of each aerosol, with the background aerosol
247 chemical mass fractions. Our control variable was restricted to the WRF-Chem output
248 variable, and the DA system changed the composition of each aerosol per size bin, depending
249 on the aerosol background errors.
250
251 Consistent with the set by Pang et al. (2020), aerosol water content (AWC) was not one of the
252 control variables in our GSI. Otherwise, the AWC might have increased contrary to the
253 physical constraints for the loading of hydrophilic particles, and simply as a mathematical
254 artefact. The AWC was diagnosed according to the analyzed aerosol mass concentration and
255 the background relative humidity. The hygroscopic growth was calculated using the WRF-
256 Chem code coupled with the revised GSI.
257
258 **2.2.2 Adjoint Operator for PM$_x$**
259 The adjoint operator for PM$_x$ is the gradient of the PM$_x$ concentration to the aerosol chemical
260 mass concentration per size bin:
261
262 $$\frac{\delta[PM_x]}{\delta[C_{aer,k}]}, k = 1, \dots, n_{size}$$
263 $$(2)$$
264
265 where n_{size} is the number of size bins and is equal to four in this study; [.] denotes the mass
266 concentration (μg m^{-3} for PM$_x$); $C_{aer,\,k}$ is the aerosol mass mixing ratio (μg kg^{-1}) of SO_4^{2-},
267 NH_4^+, OC, BC, and OIN at the k-th size bin. The threshold of aerosol mass mixing ratio that
268 yields the non-zero adjoint operator is 0.01 μg kg^{-1}. The PM$_{2.5}$ and PM$_{10}$ are assimilated in the

269   same way. When the fine and coarse particles are assimilated simultaneously, we assimilate
270   the concentration of $PM_{2.5}$ and the coarse particulate (PM_{10}-$PM_{2.5}$).
271
272   **2.2.3 Forward Operator for AOD in WRF-Chem**
273   We used the original forward operator in WRF-Chem for the aerosol optical parameters (Fast
274   et al., 2006). AOD is calculated as a function of wavelength according to Mie theory. The
275   columnar AOD τ is the sum of layer AOD across the n_z model layers:
276

277   $$\tau = \sum_{z=1}^{n_z} \tau_z = \sum_{z=1}^{n_z} \sum_{k=1}^{n_{size}} e_{ext,z,k} \cdot n_{z,k} \cdot H_z$$

278   (3)
279
280   where $e_{ext,z,k}$ is the extinction cross section of a single mixing particle in the k-th size bin at the
281   z-th model layer, $n_{z,k}$ is the aerosol number concentration, and H_z is the layer thickness. The
282   extinction cross section $e_{ext,z,k}$ of a wet particle with radius $r_{wet,z,k}$ is:
283

284   $$e_{ext,z,k} = p_{ext,z,k} \cdot \pi \cdot r_{wet,z,k}^2$$
285   (4)
286
287   where $p_{ext,z,k}$ is the extinction coefficient, given the desired mixing refractive indexes and the
288   wet particle radius. The $p_{ext,z,k}$ is attained through the Chebyshev polynomial interpolation:
289

290   $$p_{ext,z,k} = \exp\left\{ \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot c_{ext,z,k}(j) \right\}$$

291   (5)
292   where c_{ch} is the coefficient of n_{coef} order Chebyshev polynomials, $c_{ext,z,k}$ is the polynomial
293   value for the extinction efficiency of the particle, which is an internal mixture of all aerosol
294   compositions (i.e., the control variables plus nitrate, chlorine, sodium, and AWC). The radius
295   in the AOD subroutine code is in a logarithmic transform to handle the broad particle size
296   range from 0.039 μm to 10 μm. The exponential function in Eq. (5) transforms the logarithm
297   radius back to the normal radius. The aerosol number concentration $n_{z,k}$ and the aerosol dry
298   (wet) mass concentration $m_{i,z,k}$ have a linkage through the dry (wet) particle radius $r_{dry,z,k}$
299   ($r_{wet,z,k}$) and the density ρ_i of each aerosol chemical composition:
300

301   $$n_{z,k} = \sum_{i}^{n_{wet\_aer}} \frac{m_{i,z,k}}{\rho_i} \cdot \frac{3}{4\pi \cdot r_{wet,z,k}^3} = \sum_{i}^{n_{dry\_aer}} \frac{m_{i,z,k}}{\rho_i} \cdot \frac{3}{4\pi \cdot r_{dry,z,k}^3}$$

302   (6)
303
304   **2.2.4 Adjoint Operator Developed for AOD**
305   As per the forward operator in Eq. (3) in WRF-Chem, we developed the adjoint operator for
306   AOD, which requires the derivative of τ in Eq. (3) to the mass concentration, $m_{i,z,k}$:

307

308 $$\frac{\delta\tau}{\delta m_{i,z,k}} = \frac{\delta\tau_z}{\delta m_{i,z,k}} = \frac{\delta e_{ext,z,k} \cdot n_{z,k} \cdot H_z}{\delta m_{i,z,k}} + \frac{e_{ext,z,k} \cdot \delta n_{z,k} \cdot H_z}{\delta m_{i,z,k}} + \frac{e_{ext,z,k} \cdot n_{z,k} \cdot \delta H_z}{\delta m_{i,z,k}}$$

309 (7)

310

311 The first term on the righthand side of Eq. (7) indicates the change in AOD as the perturbation

312 of extinction cross section. According to Eq. (4), considering that the particle radius is

313 constant, $\delta e_{ext,z,k}$ is represented as:

314

315 $$\delta e_{ext,z,k} = \delta p_{ext,z,k} \cdot \pi \cdot r_{wet,z,k}^2$$

316 (8)

317

318 where, $\delta ch(j) = 0$ assuming that the particle radius is constant. Equation (8) is expanded with

319 the derivative of Eq. (5):

320

321 $$\delta p_{ext,z,k} = p_{ext,z,k} \cdot \{\sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot \delta c_{ext,z,k}(j)\}$$

322 (9)

323 By expanding $\delta c_{ext,z,k}$ in Eq. (9), we have:

324

325 $$\delta c_{ext,z,k}(j) = \delta w_{00} \cdot E_{ext,00}(j) + \delta w_{01} \cdot E_{ext,01}(j) + \delta w_{10} \cdot E_{ext,10}(j) + \delta w_{11} \cdot E_{ext,11}(j)$$

326 (10)

327

328 where the four parameters of E_{ext} indicate the extinction efficiencies in the Mie table

329 surrounding the point with the desired mixing refractive indexes, and the wet particle radius.

330 The changes in interpolation weights δw are determined as:

331

332 $\delta w_{00} = (v-1)\delta u + (u-1)\delta v \qquad \delta w_{01} = (1-v)\delta u - u\delta v$

333 $\delta w_{10} = (1-u)\delta v - v\delta u \qquad \delta w_{11} = u\delta v + v\delta u$

334 (11)

335

336 where

337

338 $u = \dfrac{R_{mix} - R_{low}}{R_{up} - R_{low}} \qquad \delta u = \dfrac{\delta R_{mix}}{R_{up} - R_{low}}$

339 $v = \dfrac{I_{mix} - I_{low}}{I_{up} - I_{low}} \qquad \delta v = \dfrac{\delta I_{mix}}{I_{up} - I_{low}}$

340 (12)

341

342 In Eq. (12), R_{mix} and I_{mix} are the aerosol volume-weighted mean real and imaginary refractive

343 indices, respectively. R_{up} (I_{up}) and R_{low} (I_{low}) are the nearest upper and lower limits for R_{mix}

344 (I_{mix}) in the Mie table. Considering $V_{wet,z,k}$ is the volume of all aerosol masses, the real and

345 imaginary indices and their derivatives are:

346

347 $$R_{mix,z,k} = \sum_{i}^{n_{wet\_aer}} R_i \cdot \frac{m_{i,z,k}}{\rho_i \cdot V_{wet,z,k}} \qquad \delta R_{mix,z,k} = \frac{R_i}{\rho_i \cdot V_{wet,z,k}} \cdot \delta m_{i,z,k}$$

348 $$I_{mix,z,k} = \sum_{i}^{n_{wet\_aer}} I_i \cdot \frac{m_{i,z,k}}{\rho_i \cdot V_{wet,z,k}} \qquad \delta I_{mix,z,k} = \frac{I_i}{\rho_i \cdot V_{wet,z,k}} \cdot \delta m_{i,z,k}$$

349   (13)
350
351 where

352 $$V_{wet,z,k} = \sum_{i}^{n_{wet\_aer}} \frac{m_{i,z,k}}{\rho_i}$$

353   (14)
354
355 Put Eq. (10), Eq. (11) into Eq. (9) leads to:
356
357 $$\delta p_{z,k} = [(v-1)\alpha_{00} + (1-v)\alpha_{01} - v\alpha_{10} + v\alpha_{11}]\delta u +$$
358 $$[(u-1)\alpha_{00} - u\alpha_{01} + (1-u)\alpha_{10} + u\alpha_{11}]\delta v$$
359   (15)
360 where

361 $$\alpha_{00} = p_{ext,z,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{ext,00}(j) \qquad \alpha_{01} = p_{ext,z,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{ext,01}(j)$$

362 $$\alpha_{10} = p_{ext,z,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{ext,10}(j) \qquad \alpha_{11} = p_{ext,z,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{ext,11}(j)$$

363   (16)
364
365 The first term on the righthand side of Eq. (7) is determined using Eq. (8) and Eq. (15). The
366 second term on the righthand side of Eq. (7) indicates the linkage of the aerosol number and
367 mass concentrations. It is the derivative of Eq. (6) by assuming a constant radius:
368

369 $$\delta n_{z,k} = \frac{3 \cdot \delta m_{i,z,k}}{4\pi \cdot r_{dyr,z,k}^3 \cdot \rho_i}$$

370   (17)
371
372 The third term on the righthand side of Eq. (7) contains the derivative of the layer thickness to
373 the concentrations in this layer. This indicates that the light attenuation length based on per
374 unit concentration, which can be intuitively represented by the ratio of layer thickness to the
375 aerosol mass concentration in this layer. Putting Eq. (8) and Eq. (17) into Eq. (7), we have the
376 original formula of the adjoint operator for AOD for the aerosol mass concentration:
377
378

379 $\dfrac{\delta\tau}{\delta m_{i,z,k}} = \dfrac{\delta\tau_z}{\delta m_{i,z,k}} = \dfrac{\delta e_{ext,z,k}\cdot n_{z,k}\cdot H_z}{\delta m_{i,z,k}} + \dfrac{e_{ext,z,k}\cdot \delta n_{z,k}\cdot H_z}{\delta m_{i,z,k}} + \dfrac{e_{ext,z,k}\cdot n_{z,k}\cdot \delta H_z}{\delta m_{i,z,k}} =$

380 $\{[(v-1)\alpha_{00} + (1-v)\alpha_{01} - v\alpha_{10} + v\alpha_{11}]\cdot \dfrac{\pi\cdot r_{wet,z,k}^2\cdot R_i\cdot n_{z,k}\cdot H_z}{\rho_i\cdot V_{z,k}\cdot(R_{up,z,k}-R_{low,z,k})} +$

381 $[(u-1)\alpha_{00} - u\alpha_{01} + (1-u)\alpha_{10} + u\alpha_{11}]\cdot \dfrac{\pi\cdot r_{wet,z,k}^2\cdot I_i\cdot n_{z,k}\cdot H_z}{\rho_i\cdot V_{z,k}\cdot(I_{up,z,k}-I_{low,z,k})} +$

382 $\dfrac{3e_{ext,z,k}\cdot H_z}{4\pi\cdot r_{dry,z,k}^3\cdot \rho_i} + \dfrac{e_{ext,z,k}\cdot n_{z,k}\cdot H_z}{m_{i,z,k}}\}\cdot \beta$

383

384 (18)

385

386 where β is the factor that changes the unit of mass from µg kg^{-1} to µg m^{-3}. The last righthand
387 term in Eq. (18) may not have a quick convergence in the DA outer loops because the aerosol
388 mass concentration $m_{i,z,k}$ in the denominator often has a low bias, which introduces an error
389 into the adjoint operator. The error is amplified by the layer thickness H_z in the numerator.
390 Thus, the adjoint operator of Eq. (18) cannot lead to a stable analysis. For this reason, we
391 changed the adjoint operator to account for the columnar mean aerosol extinction coefficient
392 which is described as follows:

393

394 $\dfrac{\delta\overline{(e_{ext}\cdot n)}}{\delta m_{i,z,k}} = \dfrac{H_z}{\sum H_z}\cdot \dfrac{\delta(e_{ext,z,k}\cdot n_{z,k})}{\delta m_{i,z,k}} = \dfrac{H_z}{\sum H_z}\cdot[\dfrac{\delta e_{ext,z,k}\cdot n_{z,k}}{\delta m_{i,z,k}} + \dfrac{e_{ext,z,k}\cdot \delta n_{z,k}}{\delta m_{i,z,k}}] =$

395 $\{[(v-1)\alpha_{00} + (1-v)\alpha_{01} - v\alpha_{10} + v\alpha_{11}]\cdot \dfrac{\pi\cdot r_{wet,z,k}^2\cdot R_i\cdot n_{z,k}}{\rho_i\cdot V_{z,k}\cdot(R_{up,z,k}-R_{low,z,k})} +$

396 $[(u-1)\alpha_{00} - u\alpha_{01} + (1-u)\alpha_{10} + u\alpha_{11}]\cdot \dfrac{\pi\cdot r_{wet,z,k}^2\cdot I_i\cdot n_{z,k}}{\rho_i\cdot V_{z,k}\cdot(I_{up,z,k}-I_{low,z,k})} +$

397 $\dfrac{3e_{ext,z,k}}{4\pi\cdot r_{dry,z,k}^3\cdot \rho_i}\}\cdot \beta\cdot \dfrac{H_z}{\sum H_z}$

398

399 (19)

400

401 In Eq. (19), the operator is based on the extinction coefficient at each layer, weighted by the
402 layer thickness normalized to the total model layer thickness. Correspondingly, the AOD
403 observations and AOD observation error are divided by the total layer thickness at the
404 observation location. Equation (19) is the final adjoint operator for AOD DA in this study.

405

406 **2.2.5 Adjoint Operator Developed for Surface Aerosol Attenuation Coefficients**
407 The aerosol scattering and absorption coefficients measured by the nephelometer and
408 aethalometer, respectively, are similar to the aerosol extinction coefficient at the surface in
409 Eq. (19). Neither of the two coefficients address the layer thickness. The adjoint operator for
410 the aerosol scattering coefficient measured by nephelometer is described as follows:

411

413
$$\frac{\delta(e_{sca,1,k} \cdot n_{1,k})}{\delta m_{i,1,k}} = \{[(v-1)\alpha_{00} + (1-v)\alpha_{01} - v\alpha_{10} + v\alpha_{11}] \cdot \frac{\pi \cdot r_{wet,1,k}^2 \cdot R_i \cdot n_{1,k}}{\rho_i \cdot V_{1,k} \cdot (R_{up,1,k} - R_{low,1,k})}$$

414
$$+ \frac{3e_{sca,1,k}}{4\pi \cdot r_{dry,1,k}^3 \cdot \rho_i}\} \cdot \beta$$

412   (20)

415

416
$$\alpha_{00} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{sca,00}(j) \qquad \alpha_{01} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{sca,01}(j)$$

417
$$\alpha_{10} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{sca,10}(j) \qquad \alpha_{11} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{sca,11}(j)$$

418
419   (21)
420
421   The adjoint operator for the aerosol absorption coefficient measured by aethalometer is
422

424
$$\frac{\delta(e_{abs,1,k} \cdot n_{1,k})}{\delta m_{i,1,k}} = \{[(u-1)\alpha_{00} - u\alpha_{01} + (1-u)\alpha_{10} + u\alpha_{11}] \cdot \frac{\pi \cdot r_{wet,1,k}^2 \cdot I_i \cdot n_{1,k}}{\rho_i \cdot V_{1,k} \cdot (I_{up,1,k} - I_{low,1,k})}$$

425
$$+ \frac{3e_{abs,1,k}}{4\pi \cdot r_{dry,1,k}^3 \cdot \rho_i}\} \cdot \beta$$

423   (22)

426

427
$$\alpha_{00} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{abs,00}(j) \qquad \alpha_{01} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{abs,01}(j)$$

428
$$\alpha_{10} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{abs,10}(j) \qquad \alpha_{11} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot E_{abs,11}(j)$$

429
430   (23)
431
432   where the symbols have the same meaning as before. The subscript one denotes the surface
433   layer, while. *sca* and *abs* denote "scattering" and "absorption," respectively.
434
435   As shown in the adjoint operators, the gradients of the aerosol mass concentrations rely on the
436   aerosol number concentration; meanwhile, the number concentration is estimated according to
437   the mass concentration and the particle radius. The two concentrations are intertwined in the
438   DA system, indicating the nonlinearity of the adjoint operator. This nonlinearity is handled
439   with a succeeding minimization of the cost function within the GSI. That is, the cost function
440   is first minimized with the number concentration in the background field, and the number
441   concentration is updated with the first analyzed aerosol mass concentrations. In the second
442   minimization, the number concentration assessed in the first analysis constructs a new adjoint

443   operator value, resulting in a new analysis of mass concentrations. This iterative process is
444   denoted as the "outer loop," which is repeated several times to attain the final analysis
445   (Massart et al., 2010). We used ten iterations to handle the nonlinearity in the adjoint
446   operator. The WRF-Chem AOD code is coupled into the GSI subroutine at the place of
447   invoking CRTM. The AOD and the adjoint operators of Eq. (19), Eq. (20), and Eq. (22) are
448   simultaneously determined in a single subroutine, which is cyclically invoked in the outer
449   loops within the GSI.
450
451   **2.3 Background Error Covariance (BEC)**
452   Many aerosol DA studies used the National Meteorological Center (NMC) method (Parrish
453   and Derber, 1992) to model the BEC matrix. The NMC method uses long-term archived
454   weather data that are created in the forecast cycles. It computes the statistical differences
455   between two forecasts with different leading lengths (e.g., 24 h and 48 h), but which are valid
456   at the same time. The NMC method is workable because solving global weather forecasts is
457   an initial value problem of mathematical physics. That is, a slight difference in the initial
458   atmospheric state would lead to a substantially different prediction, because of the chaos in
459   the atmosphere. However, a regional model is a boundary value problem. Meteorological
460   reanalysis data drive the regional chemistry simulation, and the driving data quality affects the
461   simulation (Giorgi and Mearns, 1999). The WRF-Chem simulations in the NMC method only
462   reflected the influences of using different initial conditions. As the model runs, the influence
463   of the initial conditions becomes weak, while the influence of lateral boundary conditions
464   always takes effect. Because the same reanalysis data drive the paring regional model
465   simulations, the following lateral boundary conditions for the simulations of the two leading-
466   lengths are similar. This leads to a limited regional model difference when using the NMC
467   method. We speculate that the NMC method cannot fully represent the model biases in
468   emission inventories and model chemistry, and it underestimates the aerosol error in WRF-
469   Chem.
470
471   Some aerosol DA studies have created background error variance using the ensemble
472   simulations by randomly disturbing model lateral boundary conditions and surface emissions
473   (Peng et al., 2017; Ma et al., 2020). The ensemble experiments better represent the model
474   error, but significantly increase the computational burden. Here, we used the standard
475   deviation of hourly aerosol concentrations in April in the background field (first guess field)
476   to represent the background error variance. The rationale of this approach is that the Tarim
477   Basin acts as a "dust reservoir" and traps dust particles for a period, before being carried long-
478   distance by wind (Fan et al., 2020). The model bias in dust dominates the model aerosol error,
479   and is correlated with the aerosol variation as the weather fluctuates. The model bias is small
480   on clear days when the aerosol concentration is low. Conversely, the bias is large when the
481   mean concentration is high: that is, on heavily-polluted days. Because the mean aerosol
482   concentration correlated positively with the aerosol variation, we used the standard deviation
483   of aerosol concentration to represent the background aerosol error. This approach was similar
484   to Sič et al. (2016), who set a percentage of the first guess field for the background error
485   variance. Our approach prioritizes DA modification of aerosols which have high background
486   mean concentrations.

487

488   We calculated the statistics of the background error, including the aerosol standard deviation
489   and the horizontal and vertical correlation length scales, using the GENerate the Background
490   Errors (GEN-BE) software (Descombes et al., 2015), based on the one-month hourly aerosol
491   concentrations in WRF-Chem. We obtained the statistics of four static BECs for the four DA
492   analysis hours (i.e., 00:00, 06:00, 12:00, and 18:00 UTC), respectively. The DA procedures
493   for the April 2019 data repeatedly use the statistics of the background error at the
494   corresponding analysis time. A usual strategy to enrich the samples of model results for
495   calculating the statistics is to gather model grid points with similar characteristics of the
496   atmosphere, referred to as "binning." The statistics are spatially averaged over the binned grid
497   points. The default strategy in the GEN_BE for GSI is latitude-binning, which creates a
498   latitude-dependent error correlation function (Figure 2a). The latitude binning is generally
499   used for latitude flow dependency and works for large and global domains (Wu et al., 2002).
500   However, we found that using the latitude-binning strategy overestimated the surface PM_x
501   concentration when assimilating aerosol optical observations. One reason for this was related
502   to the model bias in particle extinction efficiency, as discussed in Section 3.3. Another
503   plausible reason is related to the vertical profile of the background model error. The
504   maximum dust error occurred at the surface of the desert (Figure 2e) because of the local dust
505   emission sources, but the maximum error at Kashi was at the dust transporting layer above the
506   surface (Figure 2d). Owing to the vast extent of the Taklamakan Desert, the latitude-binning
507   suppressed the local error characteristics at Kashi, and led to a vertical error profile (Figure
508   2c) similar to that over the desert (Figure 2e).

509

510   For this reason, we used the standard deviation of the control variable for the OIN component
511   at each model grid to replace the latitude-binning standard deviation. The standard deviation
512   for the other compositions and the horizontal and vertical correlation length scales were
513   calculated based on the latitude-binning data. Figure 3 shows the background error statistics
514   generated by the GEN_BE software, which provided the input to the GSI. Anthropogenic
515   aerosol compositions showed vertical error profiles, greatest at the surface (Figures 3a-d). The
516   OIN component showed high background errors in the third and fourth particle sizes at the
517   transporting layer above the surface (Figure 3e). The aerosol compositions related to
518   anthropogenic emissions (i.e., sulfate, ammonium, OC, and BC, referred to here as
519   'anthropogenic aerosols') had maximum errors in the second particle size. The background
520   error for OIN composition was higher than that for anthropogenic aerosols by a factor of two
521   or three, because of the high background dust concentration in the city.

522

523   The horizontal and vertical correlation length scales determine the range of observation
524   innovations spreading from the observation locations. The horizontal influences had small
525   changes in altitude within the lowest 15 model layers (below a height of ~5 km) (Figures 3f-
526   j), indicating that the dust transport layer was well-mixed in the lower atmosphere. This deep
527   dust layer was consistent with the dust simulation by Meng et al. (2019). They showed that
528   the dust in spring was vertically mixed in a thick boundary layer to a height of 3–5 km in the
529   Tarim Basin. The vertical correlation length scales first increased from low values at the
530   surface, to high values at ~2.5 km in height (for the 8–9 layers), indicating that strong winds

531   yielded intense aerosol upward flux. The vertical correlation length scale quickly decreased
532   from the maximum value, with further increase in altitude corresponding to the large particle
533   gradient at the upper edge of the transporting layer. The latter was associated with laminar air
534   motion during the dust storm.

535

536   The background model errors were independent of particle size, which would have tended to
537   accumulate the DA modification in a single size bin that had the maximum background error
538   (e.g., the OIN in the fourth particle size). To avoid excessive accumulation of increments in a
539   single size bin, we added a one-dimensional recursive filter for the background covariances of
540   control variables across the size bins within the GSI. The inter-size bin correlation length
541   scale was two bin units, as per the setting of Saide et al. (2013).

542

Figure 2, Figure 3

543

544   **2.4 Observational Data and Errors**
545   The Dust Aerosol Observation–Kashi field campaign was performed at Kashi from
546   00:00UTC 25 March to 00:00 UTC 1 May 2019. The aerosol observations used for our DA
547   analysis included: (1) the multi-wavelength AOD measured by the sun-sky photometer
548   (Cimel CE318); (2) the multi-wavelength aerosol scattering and absorption coefficients at the
549   surface, measured with a nephelometer (Aurora 3000) and aethalometer (Magee AE-33),
550   respectively, during the campaign; and (3) the routine hourly $PM_{2.5}$ and PM_{10} observations
551   measured by the China National Environmental Monitoring Center. Please refer to Li et al.
552   (2020) for more details about the field campaign.

553

554   Table 1 summarizes the observation periods, the wavelengths of the aerosol optical data, and
555   the observation errors. The multi-wavelength data of each type of observation were
556   assimilated simultaneously. The observation errors of PM_x consisted of the measurement error
557   (e_1) and the representative error (e_2). The observation error of AOD was a constant value of
558   0.01, which was further divided by the total model layer thickness in GSI. It is difficult to
559   determine instrumental errors in nephelometers and aethalometers, and we empirically set
560   their instrumental errors to 10 Mm^{-1}, equivalent to the magnitude of the Rayleigh extinction
561   coefficient. The observational errors were uncorrelated, with **R** being a diagonal matrix.

562

Table 1

563

564   **2.5 Experimental Design**
565   The WRF-Chem simulations were configured in a two-nested domain centered at 82.9 °E,
566   41.5 °N. The coarse domain was a 120×100 (west-east × north-south) grid with a horizontal
567   resolution of 20 km that covered the Taklamakan Desert, and the fine domain was an 81×61
568   grid with a resolution of 5 km, focusing on Kashi and environs (Figure 4a). Both domains had
569   41 vertical levels extending from the surface to 50 hPa. The two domains were two-way
570   coupled. The parent domain covered the entire dust emission source, providing dust transport
571   fluxes at the lateral boundaries of the fine domain. The aerosol radiative effect was set to
572   provide feedback on the meteorology. The indirect effect of aerosols was not set in the

573 experiments. Initial and lateral boundary meteorological conditions for WRF-Chem were the
574 one-degree resolution of the National Centers for Environmental Prediction Final Analysis
575 data created by the Global Forecast System model. The meteorological lateral boundary
576 conditions for the coarse domain were updated every six hours, and were linearly interpolated
577 between the updates in WRF-Chem. We did not set the chemical boundary conditions for the
578 coarse domain. The Multiresolution Emission Inventory of China (MEIC) for 2010
579 (www.meicmodel.org) provided anthropogenic emission levels. The biogenic emission levels
580 were estimated online using the Model of Emissions of Gases and Aerosols from Nature
581 (Guenther et al., 2006). Wildfire emissions were not set in the experiments.
582
583 We conducted a one-month WRF-Chem simulation for April 2019, starting at 00:00 UTC 27
584 March and discarding the first five days for spin-up. The revised GSI system modified the
585 aerosols in the fine domain at 00:00, 06:00, 12:00, and 18:00 UTC on each day, starting from
586 00:00 UTC 1 April until the end of the month. We assimilated the observations four times a
587 day because the reanalyzed meteorological data were available for the four time slices, which
588 facilitated the model restarting from the DA analyses. The hourly PM_x observations were
589 assimilated at the exact time of analysis. The observed AOD and aerosol scattering/absorption
590 coefficients were assimilated when they fell within ±3 h, centered at the time of analysis.
591 Table 2 shows the DA experiments. The literal meanings of the experimental names denote
592 the observations that were individually or simultaneously assimilated. To study the impact of
593 DA on aerosol direct radiative forcing (ADRF), we restarted the WRF-Chem model from
594 each DA analysis, which then ran to the next analysis time. Each running performed the
595 radiation transfer calculation both with, and without aerosols, respectively. The irradiance
596 difference between the two pairing calls was aerosol radiative forcing. Section 4.2 shows the
597 DA effects on the clear-sky ADRF values.
598

Table 2, Figure 4

599
600 **3. Results**
601 **3.1 Evaluation of Control Experiment**
602 Table 2 shows the monthly mean values and correlations between the observed data and the
603 model results. The statistical values were based on the pairing data between the model results
604 and the observations. Figures 6–9 show the surface PM_x concentrations, aerosol scattering,
605 absorption coefficients, and AOD when assimilating the observations at 00:00, 06:00, 12:00,
606 and 18:00 UTC each day in April.
607
608 Kashi is in the junction between the Tian Shan Mountains to the west and the Taklamakan
609 Desert to the east (Figure 5a). In the Tarim Basin, the prevailing surface wind is easterly or
610 northeasterly, which raises dust levels and carries the particles westward (Figure 5b). An
611 intense dust storm hit the city at noon on 24 April 2019, with a peak PM_{10} concentration
612 exceeding 3,000 $\mu g\ m^{-3}$. The dust storm travelled across the northern part of the desert and
613 carried the dust particles to Kashi and the mountainous area (Figure 5c, d). A few mild dust
614 storms occurred at Kashi on April 3–5, April 8–11, and April 14–17 (Figure 6b), and the
615 maximum PM_{10} concentrations were in the range of 400–600 $\mu g m^{-3}$. The time series of $PM_{2.5}$,

616   aerosol scattering/absorption coefficient, and AOD showed patterns, similar to those for PM_{10}
617   (Figure 6).
618
619   WRF-Chem captured the main dust episodes, but significantly underestimated the aerosols at
620   Kashi (Table 2). The background monthly mean concentrations of $PM_{2.5}$ and PM_{10} were half
621   of the observed values, with a low correlation ($R < 0.3$). The simulated dust storm on 24 April
622   was a mild dust event and had a maximum PM_{10} of ~300 µg m^{-3}, one-tenth of the observed
623   value. The model lowered the aerosol scattering/absorption coefficients and AOD by 50–
624   90%.
625
626   The OIN component accounted for the model bias in PM_{10} on dusty days. Zhao et al. (2020)
627   proposed that the GOCART scheme reproduced dust emission fluxes under conditions of
628   weak wind erosion but underestimated the emissions in conditions of strong wind erosion. We
629   did not assimilate meteorology. The model bias in the surface wind introduces errors in dust
630   emission, and places bias on the number of dust particles entering the city. In the non-dust
631   days, for example, on 20 April, the model hourly $PM_{2.5}$ was 10–50% of the observed data
632   levels. The simulated anthropogenic aerosols were probably too low to be reasonable for this
633   city. The residential sector is a major source of anthropogenic emissions, including $PM_{2.5}$, BC,
634   and OC, particularly in the developing western area. The latter sector accounts for 36–82% of
635   these emissions, according to the MEIC emission inventory (Li et al., 2017). The sector is the
636   primary source of uncertainty in anthropogenic emissions inventories in China. We speculated
637   that the low bias in anthropogenic emissions could be significant for Kashi, resulting in low
638   anthropogenic aerosols in the model.
639

Figure 5

640
641   **3.2 Assimilating $PM_{2.5}$ and PM_{10} Concentrations**
642   Simultaneous assimilation of the observed PM_x (DA_PM_x) improved both the fine and coarse
643   particle concentrations, with a substantial improvement in the third and fourth particle sizes of
644   the OIN composition (Figure 11e). The analyzed monthly mean PM_{10} increased to 331.2 µg
645   m^{-3}, with a high correlation of 0.99. The analyzed monthly mean $PM_{2.5}$ was improved to 70.3
646   µg m^{-3}, although it was still lower than the observed levels, with a high correlation of 0.86.
647   The low bias in $PM_{2.5}$ was mainly in the dust storm on 24–25 April (Figure 6a). This indicates
648   that the DA system preferentially modified the coarse particle concentrations because the
649   coarse particles were assigned with a high background model error according to our BEC
650   modeling strategy. Intuitionally, this modification that mainly focused on the highest
651   concentration of coarse particles was reasonable. It decreased the model biases by raising the
652   heaviest loading aerosols. As the particle concentration increased, the aerosol scattering
653   coefficient increased to 158.9 Mm^{-1}, with a high correlation of 0.86. However, the
654   improvements in the analyzed absorption coefficients and AOD were insufficient (Figures 6d-
655   e). The analyzed absorption coefficient was 12.2 Mm^{-1}, 85% lower than observed levels, with
656   a low correlation of 0.33. The analyzed AOD showed a monthly mean value of 0.31, 56%
657   lower than observed levels, with a low correlation of 0.37.
658

659 Figure 10 shows the diurnal concentrations of PM_{10} in the analyses in April. The observed
660 PM_{10} showed a substantial variation at 18:00 UTC, the (local midnight). This substantial
661 nocturnal variation was partly owing to the dust storm that started on 24 April and ended the
662 next day. This midnight variation was also related to a nocturnal low-level jet. Ge et al.
663 (2016) pointed out that there was a nocturnal low-level jet at a height of 100–400 m, with a
664 wind speed of 4–10 m s^{-1} throughout the year in the Tarim Basin. They stressed that the low-
665 level jet broke down in the morning, transporting its momentum toward the surface, and
666 increased dust emissions. The nocturnal low-level jet increased the possibility of dust
667 particles moving towards the city at night, causing a high PM_{10} variation at 18:00 UTC. The
668 diurnal changes in the DA analyses followed the observed levels, but had higher mean values
669 (Figure 10a).
670
671 **3.3 Assimilating AOD**
672 Assimilating AOD (DA_AOD) improved the monthly mean AOD to 0.63, with a high
673 correlation of 0.99 (Figure 7e). The monthly mean $PM_{2.5}$ was improved to 85.0 µg m^{-3}, quite
674 close to the observed level, but the analyzed PM_{10} was 716.9 µg m^{-3}, which was more than
675 double the observed value. The DA system improved the AOD at the price of deteriorating
676 the data quality of surface particle concentrations, opposite to the result when assimilating
677 PM_x. Surface particle overestimations have been reported in previous studies (Liu et al., 2011;
678 Ma et al., 2020; Saide et al., 2020). Ma et al. (2020) assimilated ground-based lidars and
679 $PM_{2.5}$ simultaneously in eastern China using the WRF-Chem/DART (Data Assimilation
680 Research Testbed). They claimed that WRF-Chem underestimated the AOD and low-level
681 aerosol extinction coefficient because the model had a low bias in relative humidity, which
682 led to less AWC and lowered the single-particle extinction. As a compensation, the DA
683 system overestimated the total particle concentration to fit the observed AOD value. In the
684 arid area of Kashi, PM_{10} was strongly overestimated when assimilating AOD. We speculate
685 that WRF-Chem also lowers the dust extinction efficiency.
686
687 Table 3 shows the ratios of the AOD and aerosol scattering/absorption coefficients to the
688 surface PM_{10} concentrations in the DA_PM_x experiment. The other DA experiments yielded
689 almost similar results. This shows that the ratio of AOD to PM_{10} in the background model
690 result was only one-third of the observed levels. This low ratio indicated a model low bias in
691 particle scattering/absorption efficiency and imposed the DA system to overestimate the PM_{10}
692 to fit the observed AOD data. The low bias is related to the aerosol optical module, which is
693 based on Mie theory in WRF-Chem. First, the simulations used four-size bin particle
694 segregation. This coarse size representation aggregated many aerosols in the accumulation
695 mode. Because small particles have a strong of light attenuation capability, according to the
696 Mie theory, too many coarse particles would not effectively increase the AOD. Saide et al.
697 (2020) linked the aerosol optics to the size bin representation (from 4 to 16 bins) for hazes in
698 South Korea. They showed that WRF-Chem underestimated the dry aerosol extinction, and
699 the underestimation could be relieved when using a finer size bin than four. Okada and Kai
700 (2004) found that the dust particle radius in the Taklamakan Desert was in the range of 0.1–4
701 µm, indicating the dominant fine-mode particles in the desert. Using the four-size bin would
702 simultaneously obtain better analyses of both AOD and PM_x. Second, the dust particles are

703   irregular in shape (Okada and Kai, 2004), while the spherical particle is a common
704   assumption for the aerosol optics in the Mie theory in current models, which is an essential
705   source of uncertainty in the forward operator of WRF-Chem when the assumption of
706   spherical particles for dust fails.
707
708   Another reason for the low ratio of AOD to PM_{10} is related to our approach for modeling
709   BEC. It is important to remember that our BEC represents the possible error effects owing to
710   model bias in aerosols. The coarse particle accounts for a large mass portion of PM_x, and its
711   bias dominates the model error. However, we cannot say that this background error
712   assessment is unbiased. As our BEC gave a high background error to the coarse particle for its
713   sufficient concentration, the DA system tended to increase PM_{10}, which was not as effective
714   in increasing AOD as $PM_{2.5}$. If the background error of the coarse particle were too high, the
715   BEC would falsely lower the ratio of AOD to PM_{10} in the analysis.
716
717   To reduce the overestimate in PM_x concentrations, we set the gridded standard deviation for
718   the OIN for Kashi in place of the latitude-binning standard deviation, as discussed in Section
719   2.3. Figure 12 shows the analyzed vertical profiles of PM_x concentrations. Higher
720   concentrations were observed in the low atmosphere than at the surface. These vertical error
721   profiles decreased the surface particles and tended to increase the ratio, contrary to the effects
722   of low model bias in particle extinction efficiency and the possible high bias in the BEC
723   values of coarse particles. For the net effect of the compensation, the ratio in the analysis was
724   still almost equivalent to the background value (Table 3). That is, our tuned BEC vertical
725   profile at Kashi, to some extent canceled out the effects of other model error sources (e.g., the
726   positive bias in the coarse particle of BEC, and the low bias in extinction efficiency) but was
727   not sufficient to increase the ratio to the observed value. Finer aerosol size representation and
728   a better advanced aerosol optical calculation for dust are essential solutions.
729
730   Because the DA system overestimated the aerosol number concentration, resulting in a
731   positive bias in PM_{10}, the analyzed aerosol scattering coefficient was overestimated up to
732   280.1 Mm^{-1}, 37% higher than the observed value. In contrast, the analyzed absorption
733   coefficient was 23.1 Mm^{-1}, 72% lower than the observed value. This indicates that WRF-
734   Chem strongly underestimated the single-particle absorption efficiency, and the low bias was
735   too strong to be compensated by the overestimated aerosol number concentration.
736
737   Assimilating the AOD increased the diurnal variation in the DA analyses. There was a higher
738   increase in the concentration at noon (06:00 UTC) (Figure 10d). At the hot time of the day,
739   intense sunlight increased the light extinction by the particles. The DA system had to raise the
740   PM_{10} to fit the observed high AOD values. At dawn (00:00 UTC) or dusk (12:00 UTC), when
741   the sunlight was weak, the DA modifications were small, and the DA increases in the PM_{10}
742   fell to low levels. However, because the AOD constraint was only available in the daytime
743   and the AOD DA data were not always available as the data quality control (i.e., cloud
744   screening), assimilating AOD did not substantially increase the correlation of PM_x. The
745   analyzed PM_{10} in the DA experiments still had low correlations with the observed levels
746   (R=0.33~0.35).

747

3.4 Assimilating Aerosol Scattering Coefficient

748 **3.4 Assimilating Aerosol Scattering Coefficient**
749 Assimilating the aerosol scattering coefficient (DA_Esca) yielded overall analyses similar to
750 the phenomenon in DA_AOD. The aerosol scattering coefficient was reasonably good in the
751 analysis, with a monthly mean value of 244.6 Mm^{-1} and a high correlation of 0.97. The
752 analyzed monthly mean AOD was 0.57, better than the AOD of 0.31 when assimilating PM_x.
753 However, the surface particle concentrations were overestimated (i.e., positive biases by 32%
754 for $PM_{2.5}$, and 84% for PM_{10}), with a substantial increase in the coarse particle of OIN.
755 Overestimations appeared during the mild dust episodes (Figure 6b). This again indicated that
756 WRF-Chem underestimated the particle scattering efficiency, which was represented by the
757 ratio of the scattering coefficient to PM_{10} (Table 3). The DA system thus overfitted the PM_x
758 concentration to approach the observed scattering coefficient. The diurnal PM_{10} in the
759 analysis was similar to the assimilation of PM_x, showing a maximum improvement and a
760 robust nocturnal variation at 18:00 UTC. Assimilating the scattering coefficient failed to
761 improve the absorption coefficient. The monthly mean absorption coefficient was 19.1 Mm^{-1},
762 77% lower than the observed value.

763

764 **3.5 Assimilating Aerosol Absorption Coefficient**
765 In contrast to the above results, assimilating the absorption coefficient (DA_Eabs) degraded
766 all the analyses other than the absorption coefficient itself. The analyses showed substantial
767 daily variations, and strong positive biases appeared in the dust episodes (Figure 7). The
768 $PM_{2.5}$ was overestimated by a factor of four, and the PM_{10} was overestimated by a factor of
769 six. The increases occurred each hour and enlarged the diurnal variation of PM_{10} (Figure 10c).
770 The maximum increase in the mean value was at 06:00 UTC, also because of the strong
771 noontime heating in the model. As the particle concentration increased, the aerosol scattering
772 coefficient was overfitted to 849.0 Mm^{-1}, higher than the observed levels by a factor of four.
773 The monthly mean AOD was improbably up to 1.95. The improvement of the absorption
774 coefficient (which was 65.1 Mm^{-1}) was insufficient, and was 21% lower than the observed
775 levels.

776

777 Unlike DA_AOD and DA_Esca, assimilating the absorption coefficient cannot increase the
778 absorption data at the cost of PM_{10} overestimation. This DA failure in assimilating the
779 absorption coefficient indicates the model biases in the representation of the particle mixture
780 and the other aborting particles (e.g., brown carbon and aged dust). With respect to the current
781 model, this failure is related to the aerosol absorption represented in WRF-Chem. The leading
782 absorption aerosol in WRF-Chem is BC. The BC particle in the second size (0.156–0.625 μm)
783 had the maximum absorption, according to Mie theory, and had the maximum DA
784 modifications in the second-size bin (Figure 11d). However, because the BC had a small
785 background concentration, the BC showed a small DA improvement and had small effects on
786 increasing the particle absorption. Meanwhile, the coarse dust particle concentration was
787 primarily increased, but the dust particles did not have a strong absorption as BC. As a result,
788 the model lowered the ratio of the absorption coefficient of PM_{10} by an order of magnitude
789 (Table 3). To fit the observed absorption coefficient, the DA system dramatically

790   overestimated the particle concentrations, aerosol scattering coefficient, and AOD, but the
791   analyzed absorption coefficient was still underestimated.
792

793   **3.6 Assimilating Multi-source Observations**
794   Assimilating an individual observation improves the corresponding model parameter (i.e.,
795    $PM_{2.5}$, PM_{10}, Esca, Eabs, and AOD) but may worsen other parameters. The reasons for the
796   inconsistent improvements are relevant to the aerosol model itself. These are: (1) the model
797   parameters have opposite signs in biases (e.g., one model parameter has a positive bias while
798   another has a negative bias); (2) the model biases have vast differences in magnitude (e.g., a
799   good fit of a parameter may lead to another's overfit) and the different biases in magnitude
800   cannot be reconciled through the adjoint operator because the forward operator is inaccurate
801   (e.g., lower particle extinction efficiency). Therefore, it may not always lead to a better
802   analysis when assimilating one type of observation. Simultaneous assimilation of the multi-
803   source observations imposes more definite constraints on the DA system and helps to
804   eliminate significant model biases.
805

806   In our case, simultaneous assimilation of the scattering and absorption coefficients
807   (DA_Esca_Eabs) resulted in the analyses when assimilating the scattering coefficient alone
808   (DA_Esca), and the inferior analysis in DA_Eabs vanished. This was because incorporating
809   the scattering coefficient constrained the aerosol number concentrations, which also benefited
810   from incorporating the observed absorption coefficient. Compared with the analysis
811   assimilating the PM_x alone (DA_PM_x), assimilating the two aerosol attenuation coefficients
812   (DA_Esca_Eabs) better reproduced the AOD, but overestimated the surface particle
813   concentrations. In Figures 8–9, there were extremely high values on 28 April 2019, because
814   the scattering coefficient was missing at that time, during which the DA system assimilated
815   the absorption coefficient alone and worsened the analysis again. Simultaneous assimilation
816   of the surface particle concentration and the two aerosol attenuation coefficients
817   (DA_PM_x_Esca_Eabs) improved these three assimilated parameters, but still gave a notable
818   low bias in AOD, 41% lower than the observed levels. Simultaneous assimilation of PM_x and
819   AOD (DA_PM_x_AOD) gave the best overall DA results, in which all the analyses except the
820   absorption coefficient were not significantly different in the month mean values from the
821   observations. The analyses between DA_PM_x_AOD and DA_PM_x_Esca_Eabs were
822   comparable, except that the former additionally increased AOD better. Simultaneous
823   assimilation of all observations (DA_PM_x_Esca_Eabs_AOD) did not substantially improve
824   the analyses when compared with DA_PM_x_AOD because the surface coefficients, and AOD
825   had overlapped information of the light attenuation. A redundant information source did not
826   introduce extra constraints on the DA system. All the DA experiments failed to improve the
827   aerosol absorption coefficient, which always showed strong, low biases (> 76%) and low
828   correlations (< 0.5) in the analyses, implying room for improvement of our DA system.
829

Table 3, 4; Figure 6, 7, 8, 9, 10, 11

830

831   **3.7 Vertical Profiles of Aerosol Concentrations**

832   Figure 12 shows the vertical concentration profiles of $PM_{2.5}$ and PM_{10}. The DA system
833   increased the aerosol concentrations up to a height of 4 km, which is consistent with previous
834   studies on the Taklamakan Desert. Meng et al. (2019) simulated a deep dust layer thickness in
835   spring, with a depth of 3–5 km. Ge et al. (2014) analyzed the Cloud-Aerosol Lidar Orthogonal
836   Polarization data from to 2006–2012 in the desert. They showed that dust could be lifted up to
837   5 km above the Tarim Basin, and even higher along the northern slope of the Tibetan Plateau.
838   Among our DA experiments, the analyzed PM_x in the lower atmosphere followed PM_x at the
839   surface. The vertical PM_{10} concentration increased quickly in the lowest three model layers
840   and maintained high values at heights of less than 3 km. This vertical profile corresponded
841   well to the background vertical error profile (Figure 3e), reflecting the deep dust transporting
842   layer. The $PM_{2.5}$ vertical profiles of showed a rapid reduction with an increase in altitude. The
843   figure clearly shows that DA_PMx improved the PM_{10} better than $PM_{2.5}$, whereas DA_AOD
844   preferentially adjusted the coarse particles and overestimated the PM_{10}. DA_PMx._AOD
845   provided the best balance between the adjustments of $PM_{2.5}$ and PM_{10}.
846

Figure 12

847
848   **4. Discussion**
849   **4.1 DA Impact on Aerosol Chemical Composition**
850   The maximum concentrations of sulfate, ammonium, BC, and OC in April were 4.1, 1.5, 0.5,
851   and 1.3 $\mu g\ m^{-3}$, respectively, in the background model data. Although a careful evaluation is
852   difficult because of the lack of aerosol chemical measurements, we speculated that the
853   aerosols (other than OIN) were considerably low. Anthropogenic emissions might be biased
854   for this city. The sources of emissions in residential/developing areas are principally
855   anthropogenic; yet the residential emission factor for the emission inventory compilation is
856   highly uncertain compared with the emission factors of power plants, industrial plants, and
857   vehicles (Li et al., 2017). Chlorine and sodium are selected to represent sea-salt aerosols in
858   WRF-Chem, yet the two concentrations were at very low concentrations in the model at
859   Kashi. This was despite the fact that the Taklamakan Desert had many atmospheric halite
860   particles, which were Cl- and Na-rich and accounted for 10% of the total particles in the
861   desert (Okada and Kai, 2004).
862
863   For control variable design, our DA system modifies the chemical composition of each
864   aerosol according to the BEC values. However, all the DA experiments showed that the PM_{10}
865   chemical fractions remain close to their background values (Figure 13). The low biases cannot
866   be improved via DA because the aerosol chemical measurements were not available. In
867   addition, the differences between DA_Esca and DA_Esca_Eabs were quite small (Figure 13c,
868   e), indicating that assimilating the aerosol absorption coefficient did not enhance particle
869   absorption (19.1^{-1} vs. 20.0 Mm^{-1} in Table 3) in our system. The assimilation of the aerosol
870   absorption coefficient alone (DA_Eabs) increased the percentage of BC to 7.1%, which was
871   slightly higher than the 6.5% background BC. The first reason for the small changes in the
872   aerosol chemical proportions is that the scattering aerosols (i.e., sulfate, nitrate, and
873   ammonium), use the same refractive index and hygroscopicity parameter in WRF-Chem.
874   Therefore, the AOD had virtually the same sensitivity to composition of each aerosol, and

875   assigned comparable modification to each composition. When assimilating the total quantities
876   of aerosols (e.g., $PM_{2.5}$, PM_{10}, and AOD), it is difficult to distinguish different aerosol
877   chemical contributions. Secondly, DA_Eabs increased the aerosol number concentration;
878   meanwhile, the rising number concentration increased the scattering aerosols, which
879   prevented a substantial rise in the BC fraction. Third, the concentrations of anthropogenic
880   aerosols were lower than anticipated. They had comparable low background errors and could
881   not be distinguished because of their small differences in BEC values. Overall, it seems that
882   differences in aerosol chemical composition from assimilating the aerosol optical data are
883   smaller than the difference in model setting (e.g., using other aerosol chemistry mechanisms,
884   or using finer aerosol size bins). The assimilation of the total aerosol quantities cannot
885   eliminate the intrinsic bias in aerosol composition. Thus, accurate aerosol chemistry and
886   optical modules are crucial to attain a better background aerosol chemical data for DA
887   analysis (Saide et al., 2020).
888

Figure 13

889
890   **4.2 DA Impact on Aerosol Direct Radiative Forcing**
891   Table 4 shows the instantaneous clear-sky ADRF in the background data and the analyses of
892   DA_PM_x and DA_PM_x_AOD. After the analyses, the DA effect (various DA frequencies for
893   assimilating AOD and the surface particle concentrations) gradually faded away after
894   restarting the model run. We therefore focused on the instantaneous radiative forcing values
895   one hour after assimilating AOD data. This ensured that the comparison was based on similar
896   analysis times and showed effective DA effects. As the dust was the predominant component,
897   the ADRF in this section was closely equivalent to the dust radiative forcing.
898
899   Dust redistributes the energy between the land and the atmosphere. The atmosphere gains
900   more shortwave energy as the dust particle absorption; the warming atmosphere also emits
901   more longwave energy as it absorbs shortwave energy. The change in energy budget at the
902   surface is correspondingly the opposite of that in the atmosphere. As shown in Table 4, the
903   enhancements in surface cooling forces were slightly stronger than the atmospheric warming
904   forcings. The differences between the surface forcing and atmospheric forcing indicate the
905   ADRF at the top of the atmosphere (TOA). The TOA ADRF when assimilating the surface
906   particle concentrations was enhanced by 12% in the shortwave, 83% in the longwave, and 6%
907   in the net forcing values, and enhanced by 40%, 55%, and 38%, respectively, when
908   assimilating the AOD. Apparently, assimilating PM_x alone is not sufficient to accurately
909   estimate the ADRF value. At Kashi, the total net clear-sky ADRF with assimilating surface
910   particles and AOD were –10.5 Wm^{-2} at the TOA, +19.5 Wm^{-2} within the atmosphere, and –
911   30.0 Wm^{-2} at the surface, respectively, enhanced by 46%, 153%, and 100% respectively,
912   compared to the background ADRF values. Because the AOD observation is only
913   sporadically available because of cloud screening in retrieval data, the DA experiments still
914   cannot eliminate the low bias in AOD in WRF-Chem. The ADRF values in the DA
915   experiments are still likely to be lower than the plausible aerosol radiative forcing at Kashi.
916
917   **5. Conclusions**

918   This study described our revised GSI DA system for assimilating aerosol observed data for
919   the four-size bin sectional MOSAIC aerosol mechanism in WRF-Chem. The DA system has
920   new design adjoint operators for the multi-wavelength AOD, aerosol scattering, and
921   absorption coefficients measured by the sun-sky radiometer, nephelometer, and aethalometer,
922   respectively. We examined the DA system for Kashi city in northwestern China by
923   assimilating the multi-wavelength aerosol optical measurements gathered by the Dust Aerosol
924   Observation–Kashi field campaign of April 2019 and the concurrent hourly measurements of
925   surface $PM_{2.5}$ and PM_{10} concentrations from the local environmental monitoring sites.

926

927   Our DA system includes two main aspects. Firstly, the control variable is the aerosol chemical
928   composition per size bin corresponding to the WRF-Chem output data. This design allows the
929   modification of the composition of each aerosol, based on their background error covariances.
930   The number of control variables could be reduced by intentionally excluding a few aerosol
931   compositions in a specific case, if these compositions had low concentrations. Second, the DA
932   system incorporates the observed AOD by assimilating the column mean aerosol extinction
933   coefficient. This transfer avoids handling sensitivity from light attenuation length to the
934   aerosol mass concentration in the adjoint operator, which is difficult to accurately estimate
935   and introduces significant errors in the operator. The adjoint operator for AOD has two
936   variants that incorporate nephelometer and aethalometer measurements.

937

938   The most abundant aerosol at Kashi in April 2019 was dust. The WRF-Chem model captured
939   the main dust episodes, but only simulated half the monthly mean concentrations of $PM_{2.5}$ and
940   PM_{10}. Furthermore, the model failed to capture the peak concentrations from a dust storm on
941   24 April. The aerosol scattering/absorption coefficients and AOD in the background data
942   showed strong low biases and weak correlations with the observed levels. The DA systems
943   did, however, effectively assimilate the surface particle concentrations, aerosol scattering
944   coefficients, and AOD. Some deficiencies in the DA analysis were related to the forward
945   model bias in transferring the aerosol mass concentrations to the aerosol optical parameter.
946   Simultaneous assimilation of the $PM_{2.5}$ and PM_{10} concentrations improved the model aerosol
947   concentrations, with significant increases in the coarse particles; meanwhile, the analyzed
948   AOD was 56% lower than observed levels. The assimilation of AOD significantly improved
949   the AOD but overestimated the surface PM_{10} concentration by a factor of at least two.
950   Assimilating the aerosol scattering coefficient improved the scattering coefficient in the
951   analysis but overestimated the surface PM_{10} concentration by 84%. It therefore seems that
952   WRF-Chem underestimated the particle extinction efficiency. As a compensation, the DA
953   system overestimated the aerosol concentration to fit the observed optical values, yielding
954   overly high particle concentrations.

955

956   A notable problem was the assimilation of the absorption coefficient, which greatly
957   overestimated the monthly mean values by a factor of at least four in the model parameters
958   and yielded overly strong daily variations for the parameters. The aerosol absorption
959   coefficient was improved but was still 21% lower than observed values. This defect was also
960   apparent in the low particle absorption efficiency of WRF-Chem. The biases of the model in
961   aerosol particle mixture and aged dust as well as the "missing" absorption of brown carbon,

Atmospheric
Chemistry
and Physics
Discussions

Open Access
EGU

962 accounted for the bias in absorption efficiency, which would have worsened the DA analysis
963 when assimilating the absorption coefficient.

964

965 Simultaneous assimilation of the multi-source observations imposes a more definite constraint
966 and helps improve model parameters. Simultaneously assimilating the scattering and
967 absorption coefficients eliminated the defect of assimilating the absorption coefficient. It also
968 provided comparable improvements for assimilating the surface particles and AOD; the latter
969 additionally improved the AOD analysis. The most effective DA is the simultaneous
970 assimilation of surface particle concentration and AOD, which provides the best overall DA
971 analysis.

972

973 Our design of control variables allowed the DA system to adjust the aerosol chemical
974 compositions individually. However, the analyzed anthropogenic aerosol chemical fractions
975 were almost equivalent to the background chemical fractions. The reason is that the
976 hydrophilic aerosols have equivalent or comparable refractive indices and hygroscopic
977 parameters in the forward operator; they therefore have comparable adjoint operator values
978 when assimilating the aerosol optical data. It may be possible to separate the chemical
979 compositions based on their background errors. In our case, the anthropogenic aerosols were
980 unrealistically low at Kashi, probably owing to the low biases in the anthropogenic emissions.
981 The low background concentrations led to low background errors and hence few increments
982 for all chemical compositions. As a result, the chemical fractions of the anthropogenic
983 aerosols remained close to their background values.

984

985 When assimilating surface particles and AOD, the instantaneous clear-sky ADRF at Kashi
986 were –10.5 Wm^{-2} at the TOA, +19.5 Wm^{-2} within the atmosphere, and –30.0 Wm^{-2} at the
987 surface, respectively. Since the DA analyses still lowered the AOD value, the aerosol
988 radiative forcing values assimilating the observations were also underestimated.

989

990 The limitations that necessitate further research include:
991   (1) The binning strategy. The desired strategy should link the circulation flow and
992 particle emission sources. A better hybrid DA coupled with the ensemble Kalman filter will
993 be more effective for estimating the aerosol background error.
994   (2) The observational error. This could be elaborated further. The PM_{10} included the
995 anthropogenic coarse particles, which should be separated from the dust originating from the
996 desert (Jin et al., 2019). We set the observation errors for PM_x and AOD to the conventional
997 values. The observational errors of the nephelometer and aethalometer were slightly arbitrary
998 in this study, necessitating further consideration.
999   (3) The adjoint operator. This needs to be modified to assimilate the aerosol absorption
1000 coefficient or absorption AOD.
1001   (4) The DA system. Our revised DA system was based on four-size bin MOSAIC
1002 aerosols, but it can be extended to work with eight-size bin MOSAIC aerosols in WRF-Chem.
1003 When assimilating aerosol optical data, the DA quality is strongly dependent on the forward
1004 model. The responses of our DA analysis to the bias and uncertainty in the forward aerosol
1005 optical model in WRF-Chem thus need further investigation.

Author contributions

WC developed the DA system, preformed the analyses and wrote the paper. ZL led the field campaign and revised the paper. YZ and KL implemented the observations and the data quality control. YZ helped to design the new adjoint operator. JC verified the DA system.

Competing interests

The authors declare that they have no conflict of interest.

Code/Data availability

The official GSI code is available at https://dtcenter.org/community-code/gridpoint-statistical-interpolation-gsi/download. The aerosol measurements at Kashi belong to the Sun-sky radiometer Observation NETwork (SONET) which is accessible at http://www.sonet.ac.cn/en/index.php.

References

Bannister, R. N.: A review of operational methods of variational and ensemble-variational data assimilation, Q.J.R. Meteorol. Soc., 143, 607-633, doi:10.1002/qj.2982, 2017.

Bao, Y., Zhu, L, Guan, Q., Guan, Y., Lu, Q., Petropoulos, G. P., Che H., Ali, g., Dong, Y., Tang, Z., Gu, Y., Tang, W., and Hou, Y.: Assessing the impact of Chinese FY-3/MERSI AOD data assimilation on air quality forecasts: sand dust events in northeast China, Atmos. Environ., 205, 78-89, doi:10.1016/j.atmosenv.2019.02.026, 2019.

Calil, P. H. R., Doney, S. C., Yumimoto, K., Eguchi, K., and Takemura, T.: Episodic upwelling and dust deposition as bloom triggers in low-nutrient, low-chlorophyll regions, J. Geophys. Res., 116, C06030, doi:10.1029/2010jc006704, 2011.

Chen, D., Liu, Z., Schwartz, C. S., Lin, H.-C., Cetola, J. D., Gu, Y., and Xue, L.: The impact of aerosol optical depth assimilation on aerosol forecasts and radiative effects during a wild fire event over the United States, Geosci. Model Dev., 7, 2709-2715, doi:10.5194/gmd-7-2709-2014, 2014.

Chen, L., Gao, Y., Zhang, M., Fu, J. S., Zhu, J., Liao, H., Li, J., Huang, K., Ge, B., Wang, X., Lam, Y. F., Lin, C.-Y., Itahashi, S., Nagashima, T., Kajino, M., Yamaji, K., Wang, Z., and Kurokawa, J.-i.: MICS-Asia III: multi-model comparison and evaluation of aerosol over East Asia, Atmos. Chem. Phys., 19, 11911-11937, doi:10.5194/acp-19-11911-2019, 2019.

Chen, S.-H., and Sun, W.-Y.: A one-dimensional time dependent cloud model, J. Meteor. Soc. Japan, 80, 1, 99-118, doi:10.2151/jmsj.80.99, 2002.

1050   Cheng, X., Liu, Y., Xu, X., You, W., Zang, Z., Gao, L., Chen, Y., Su, D., and Yan, P.: Lidar
1051   data assimilation method based on CRTM and WRF-Chem models and its application in
1052   PM$_{2.5}$ forecasts in Beijing, Sci. Total Environ., 682, 541-552,
1053   doi:10.1016/j.scitotenv.2019.05.186, 2019.
1054
1055   Descombes, G., Auligné, T., Vandenberghe, F., Barker, D. M., and Barré, J.: Generalized
1056   background error covariance matrix model (GEN_BE v2.0), Geosci. Model Dev., 8, 669-696,
1057   doi:10.5194/gdm-8-669-2015, 2015.
1058
1059   Fan, J., Shang, Y., Chen, Q., Wang, S., Zhang, X., Zhang, L., Zhang, Y., Xu, X., and Jiang,
1060   P.: Investigation of the "dust reservoir effect" of the Tarim Basin using WRF-GOCART
1061   model, Arab. J. Geosci., 13, 214, doi:10.1007/s12517-020-5154-x, 2020.
1062
1063   Fast, J. D., Gustafson, W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G.,
1064   Grell, G. A. Peckham, S. E.: Evolution of ozone, particulates, and aerosol direct radiative
1065   forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol
1066   model, J. Geophys. Res.-Atmos., 111, D21305, doi:10.1029/2005jd006721, 2006.
1067
1068   Feng, S., Jiang, F., Jiang, Z., Wang, H., Cai, Z., and Zhang, L.: Impact of 3DVAR
1069   assimilation of surface PM$_{2.5}$ observations on PM$_{2.5}$ forecasts over China during wintertime,
1070   Atmos. Environ., 187, 34-49, doi:10.1016/j.atmosenv.2018.05.049, 2018.
1071
1072   Ge, J. M., Huang, J. P., Xu, C. P., Qi, Y. L., and Liu, H. Y.: Characteristics of Taklimakan
1073   dust emission and distribution: A satellite and reanalysis field perspective, J Geophys. Res.
1074   Atmos., 119, 11772-11783, doi:10.1002/2014jd022280, 2014.
1075
1076   Ge, J. M., Liu, H., Huang, J., and Fu, Q.: Taklimakan desert nocturnal low-level jet:
1077   climatology and dust activity, Atmos. Chem. Phys., 16, 7773-7783, doi:10.5194/acp-16-7773-
1078   2016, 2016.
1079
1080   Ginoux, P., Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O., and Lin, S.-J.:
1081   Sources and distributions of dust aerosols simulated with the GOCART model, J. Geophys.
1082   Res., 106, D17, 20255-20273, doi: 10.1029/2000JD000053, 2001.
1083
1084   Giorgi, F., and Mearns, L. O.: Introduction to special section: Reginal climate modeling
1085   revisited, J. Geophys. Res., 104, D6, 6335-6352, doi:10.1029/98JD02072, 1999.
1086
1087   Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and
1088   Eder, B.: Fully coupled "online" chemistry within the WRF model. Atmos. Environ., 39,
1089   6957-7975, doi:10.1016/j.atmosenv.2005.04.027, 2005.
1090
1091   Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of
1092   global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and

Atmospheric
Chemistry
and Physics
Discussions

1093   Aerosols from Nature), Atmos. Chem. Phys., 6, 3181-3210, doi:10.5194/acp-6-3181-2006,
1094   2006.
1095
1096   He, J., Zhang, Y., Wang, K., Chen, Y., Leung, L. R., Fan, J., Li, M., Zheng, B., Zhang, Q.,
1097   Duan, F., and He, K.: Multi-year application of WRF-CAM5 over East Asia-Part I:
1098   Comprehensive evaluation and formation regimes of O_3 and $PM_{2.5}$, Atmos. Environ., 165,
1099   122-142, doi:10.1016/j.amtosenv.2017.06.015, 2017.
1100
1101   Hong, S.-Y., Noh, Y., Dudhia, J.: A new vertical diffusion package with an explicit treatment
1102   of entrainment processes, Mon. Wea. Res., 134, 2318-2341, doi:10.1175/MWR3199.1, 2006.
1103
1104   Huang, J., Wang, T., Wang, W., Li, Z., and Yan, H.: Climate effects of dust aerosols over
1105   East Asian arid and semiarid regions, J. Geophys. Res., 119, 11398-11416,
1106   doi:10.1002/2014jd021796, 2014.
1107
1108   Hong, J., Mao, F., Min, Q., Pan, Z., Wang, W., Zhang, T., and Gong, W.: Improved $PM_{2.5}$
1109   predictions of WRF-Chem via the integration of Himawari-8 satellite data and ground
1110   observations, Environ. Pollut., 263, 114451, doi:10.1016/j.envpol.2020.114451, 2020.
1111
1112   Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and Collins,
1113   W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative
1114   transfer models, J. Geophys. Res., 113, D13103, doi:10.1029/2008JD009944, 2008.
1115
1116   Jia, R., Liu, Y., Chen, B., Zhang, Z., and Huang, J.: Source and transportation of summer dust
1117   over the Tibetan Plateau, Atmos. Environ., 123, 210-219,
1118   doi:10.1016/j.atmosenv.2015.10.038, 2015.
1119
1120   Jiang, Z., Liu, Z., Wang, T., Schwartz, C. S., Lin, H.-C., and Jiang, F.: Probing into the
1121   impact of 3DVAR assimilation of surface PM_{10} observations over China using process
1122   analysis, J. Geophys. Res., 118, 6738-6749, doi:10.1002/jgrd.50495, 2013.
1123
1124   Jin, J., Lin, H. X., Segers, A., Xie, Y., and Heemink, A.: Machine learning for observation
1125   bias correction with application to dust storm data assimilation, Atmos. Chem. Phys., 19,
1126   10009-10026, doi:10.5194/acp-19-10009-2019.
1127
1128   Li, L., Li, Z., Chang, W., Ou, Y., Goloub, P., Li, C., Li, K., Hu, Q., Wang, J., and Wendisch,
1129   M.: Solar radiative forcing of aerosol particles near the Taklimakan desert during the Dust
1130   Aerosol Observation-Kashi campaign in Spring 2019, Atmos. Chem. Phys. Discuss.,
1131   doi:10.5194/acp-2020-60, in review, 2020.
1132
1133   Li, M., Liu, H., Geng, G., Hong, C., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H., Man, H.,
1134   Zhang, Q., and He, K.: Anthropogenic emission inventories in China: a review, Natl. Sci.
1135   Rev., 4, 834-866, doi:10.1093/nsr/nwx150, 2017.
1136

1137 Liu, Z., Liu, Q., Lin, H.-C., Schwartz, C. S., Lee, Y.-H., and Wang, T.: Three-dimensional
1138 variational assimilation of MODIS aerosol optical depth: Implementation and application to a
1139 dust storm over East Asia, J. Geophys. Res., 116, D23206, doi:10.1029/2011JD016159, 2011.
1140
1141 Li, Z., Zang, Z., Li, Q. B., Chao, Y., Chen, D., Ye, Z., Liu, Y., and Liou, K. N.: A three-
1142 dimensional variational data assimilation system for multiple aerosol species with
1143 WRF/Chem and an application to $PM_{2.5}$ prediction, Atmos. Chem. Phys., 13, 4265-4278,
1144 doi:10.5194/acp-13-4265-2013, 2013.
1145
1146 Ma, C., Wang, T., Mizzi, A. P., Anderson, J. L., Zhuang, B., Xie, M., and Wu, R.:
1147 Multiconstituent data assimilation with WRF-Chem/DART: Potential for adjusting
1148 anthropogenic emissions and improving air quality forecasts over eastern China, J. Geophys.
1149 Res. Atmos., 124, 7393-7412, doi:10.1029/2019JD030421, 2019.
1150
1151 Ma, C., Wang, T., Jiang, Z., Wu, H., Zhao, M., Zhuang, B., Li, S., Xie, M., Li, M., Liu, J.,
1152 and Wu, R.: Importance of bias correction in data assimilation of multiple observations over
1153 eastern China using WRF-Chem/DART, J. Geophys. Res. Atmos., 125, e2019JD031465,
1154 doi:10.1029/2019JD031465, 2020.
1155
1156 Massart, S., Pajot, B., Piacentini, A., and Pannekoucke, O.: On the merits of using a 3D-
1157 FGAT assimilation scheme with an outer loop for atmospheric situations governed by
1158 transport, Mon. Weather Rev., 138, 12, 4509-4522, doi:10.1175/2010MWR3237.1, 2010.
1159
1160 Meng, L, Yang, X., Zhao, T., He, Q., Lu, H., Mamtimin, A., Huo, W., Yang, F., and Liu, C.:
1161 Modeling study on three-dimensional distribution of dust aerosols during a dust storm over
1162 the Tarim Basin, Northwest China, Atmos. Res., 218, 285-295,
1163 doi:10.1016/j.atmores.2018.12.006, 2019.
1164
1165 Okada, K., and Kai, K.: Atmospheric mineral particles collected at Qira in the Taklamakan
1166 Desert, China, Atmos. Environ., 38, 6927-6935, doi:10.1016/j.atmosenv.2004.03.078, 2004.
1167
1168 Pagowski, M., Grell, G. A., McKeen, S. A., Peckham, S. E., and Devenyi, D.: Three-
1169 dimensional variational data assimilation of ozone and fine particulate matter observations:
1170 some results using the Weather Research and Forecasting – Chemistry model and Grid-point
1171 Statistical Interpolation, Q. J. R., Meteorol. Soc., 136, 2014-2024, doi:10.1002/qj.700, 2010.
1172
1173 Pagowski, M., Liu, Z., Grell, G. A., Hu, M., Lin, H.-C., and Schwartz, C. S.: Implementation
1174 of aerosol assimilation in Gridpoint Statistical Interpolation (v.3.2) and WRF-Chem (v.3.4.1),
1175 Geosci. Model Dev., 7, 1621-1627, doi:10.5194/gmd-7-1621-2014, 2014.
1176
1177 Pang, J., Liu, Z., Wang, X., Bresch, J., Ban, J., Chen, D., and Kim, J.: Assimilating AOD
1178 retrievals from GOCI and VIIRS to forecast surface $PM_{2.5}$ episodes over eastern China,
1179 Atmos. Environ., 179, 288-304, doi:10.1016/j.atmosenv.2018.02.011, 2018.
1180

1181   Pang, J., Wang, X., Shao, M., Chen, W, and Chang, M.: Aerosol optical depth assimilation for
1182   a modal aerosol model: Implementation and application in AOD forecasts over East Asia, Sci.
1183   Total Environ., 719, 137430, doi:10.1016/j.scitotenv.2020.137430, 2020.

1185   Parrish, D. F., and Derber, J. C.: The National Meteorological Center's spectral statistical-
1186   interpolation analysis system, Mon. Weaterh Rev., 120, 1747-1763, doi: 10.1175/1520-
1187   0493(1992)120<1747:TNMCSS>2.0.CO;2, 1992.

1189   Peng, Z., Liu, Z., Chen, D., and Ban, J.: Improving $PM_{2.5}$ forecast over China by the joint
1190   adjustment of initial conditions and source emissions with an ensemble Kalman filter, Atmos.
1191   Chem. Phys., 17, 4837-4855, doi:10.5194/acp-17-4837-2017, 2017.

1193   Peng, Z., Lei, L., Liu, Z., Sun, J., Ding, A., Ban, J., Chen, D., Kou, X., and Chu, K.: The
1194   impact of multi-species surface chemical observation assimilation on air quality forecasts in
1195   China, Atmos. Chem. Phys., 18, 17387-17404, doi:10.5194/acp-18-17387-2018, 2018.

1197   Purser, R. J., Wu, W.-S., Parrish, D. F., and Roberts, N. M.: Numerical aspects of the
1198   application of recursive filters to variational statistical analysis. Part I: spatially homogeneous
1199   and isotropic gaussian covariances, Mon. Weather Rev., 131, 1524-1535, doi: 10.1175//1520-
1200   0493(2003)131<1524:NAOTAO>2.0.CO;2, 2003a.

1202   Purser, R. J., Wu, W.-S., Parrish, D. F., and Roberts, N. M.: Numerical aspects of the
1203   application of recursive filters to variational statistical analysis. Part II: spatially
1204   inhomogeneous and anisotropic general covariances, Mon. Weather Rev., 131, 1536-1548,
1205   doi: 10.1175//2543.1, 2003b.

1207   Saide, P. E., Carmichael, G. R., Liu, Z., Schwartz, C. S., Lin, H. C., da Silva, A. M., and
1208   Hyer, E.: Aerosol optical depth assimilation for a size-resolved sectional model: impacts of
1209   observationally constrained, multi-wavelength and fine mode retrievals on regional scale
1210   analyses and forecasts, Atmos. Chem. Phys., 13, 10425-10444, doi:10.5194/acp-13-10425-
1211   2013, 2013.

1213   Saide, P. E., Kim, J., Song, C. H., Choi, M., Cheng, Y., and Carmichael, G. R.: Assimilation
1214   of next generation geostationary aerosol optical depth retrievals to improve air quality
1215   simulations, Geophys. Res. Lett., 41, 9188-9196, doi:10.1002/2014GL062089, 2014.

1217   Saide, P. E., Gao, M., Lu, Z., Goldberg, D., Streets, D. G., Woo, J.-H., Beyersdorf, A., Corr,
1218   C. A., Thornhill, K. L., Anderson, B., Hair, J. W., Nehrir, A. R., Diskin, G. S., Jimenez, J. L.,
1219   Nault, B. A., Campuzano-Jost, P., Dibb, J., Heim, E.,, Lamb, K. D., Schwarz, J. P., Perring,
1220   A. E., Kim, J., Choi, M., Holben, B., Pfister, G., Hodzic, A., Carmichael, G. R., Emmons, L.,
1221   and Crawford, J. H.: Understanding and improving model representation of aerosol optical
1222   properties for a Chinese haze event measured during KORUS-AQ, Atmos. Chem. Phys., 20,
1223   6455-6478, doi:10.5194/acp-20-6455-2020, 2020.

1225   Schwartz, C. S., Liu, Z., Lin, H.-C., and McKeen, S. A.: Simultaneous three-dimensional
1226   variational assimilation of surface fine particulate patter and MODIS aerosol optical depth, J.
1227   Geophys. Res., 117, D13202, doi:10.1029/2011JD017383, 2012.
1228
1229   Sič, B., Amraoui, L. E., Piacentini, A., Marécal, V., Emili, E., Cariolle, D., Prather, M., and
1230   Attié, J.-L.: Aerosol data assimilation in the chemical transport model MOCAGE during the
1231   TRAQA/ChArMEx campaign: aerosol optical depth, Atmos. Chem. Phys., 9, 5535-5554,
1232   doi:10.5194/amt-9-5535-2016, 2016.
1233
1234   Tang, Y., Pagowski, M., Chai, T., Pan, L., Lee, P., Baker, B., Kumar, R., Monache, L .D.,
1235   Tong, D., and Kim, H.-C.: A case study of aerosol data assimilation with the Community
1236   Multi-scale Air Quality Model over the contiguous United States using 3D-Var and optimal
1237   interpolation methods, Geosci. Model Dev., 10, 4743-4758, doi:10.5194-gmd-10-4743-2017,
1238   2017.
1239
1240   Tewari, M., Chen, F., Wang, W., Dudhai, J., LeMone, M. A., Mitchell, K., Ek, M., Gayno,
1241   G., Wegiel, J., and Cuenca, R. H.: Implementation and vertification of the unified NOAH land
1242   surface model in the WRF model. 20th conference on weather analysis and forecasting/16th
1243   conference on numerical weather prediction, pp. 11-15.
1244
1245   Wang, K.-Y., Lary, D. J., Shallcross D. E., Hall, S. M., and Pyle, J. A.: A review on the use of
1246   the adjoint method in four-dimensional atmospheric-chemistry data assimilation, Q. J. R.
1247   Meteorol. Soc., 127, 2181-2204, doi:10.1002/qj.49712757616, 2001.
1248
1249   Wu, W.-S., Purser, r. J., and Parrish, D. F.: three-dimensional variational analysis with
1250   spatially inhomogeneous covariances, Mon. Weather Rev., 130, 12, 2905-2916,
1251   doi: 10.1175/1520-0493(2002)130<2905:TDVAWS>2.0.CO;2, 2002.
1252
1253   Xia, X., Min, J., Shen, F., Wang, Y., and Yang, C.: Aerosol data assimilation using data from
1254   Fengyun-3A and MODIS: application to a dust storm over East Asia in 2011, Adv. Atmos.
1255   Sci., 36, 1-14, doi:10.1007/s00376-018-8075-9, 2019a.
1256
1257   Xia, X., Min, J., Wang, Y., Shen, F., Yang, C., and Sun, Z.: Assimilating Himawari-8 AHI
1258   aerosol observations with a rapid-update data assimilation system, Atmos. Environ., 215,
1259   116866, doi:10.1016/j.atmosenv.2019.116866, 2019b.
1260
1261   Zang, Z., Li, Z., Pan, X., Hao, Z., and You, W.: Aerosol data assimilation and forecasting
1262   experiments using aircraft and surface observations during CalNex, Tellus B., 68,1, 29812,
1263   doi:10.3402/tellusb.v68.29812, 2016.
1264
1265   Zaveri, R. A., and Peters, L. K.: A new lumped structure photochemical mechanism for large-
1266   scale applications, J. Geophys. Res., 104, 30387-30415, doi:10.1029/1999JD900876, 1999.
1267

1268   Zaveri, R. A., Easter, R. C., Fast, J. D., Peters, L. K.: Model for simulating aerosol
1269   interactions and chemistry (MOSAIC), J. Geophys. Res., 113, D13204,
1270   doi:10.1029/2007JD008782, 2008.
1271
1272   Zhao, J., Ma, X., Wu, S., and Sha, T.: Dust emission and transport in Northwest China: WRF-
1273   Chem simulation and comparisons with multi-sensor observation, Atmos. Res., 241, 104978,
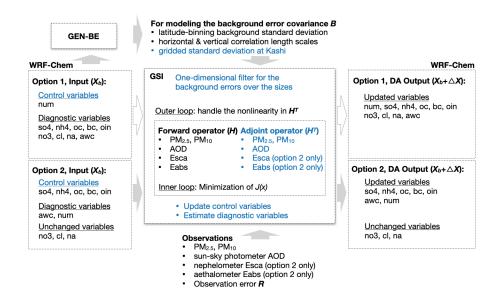1274   doi:10.1016/j.atmosres.2020.104978, 2020.

Figure 1. The workflow of aerosol DA in the revised GSI system for the sectional MOSAIC aerosols in WRF-Chem. The contents in blue are the portions we developed. The arrows in gray indicate the workflow of option 2 that we did in this study. Only option 2 can assimilate the aerosol scattering/absorption coefficients. Abbreviations: so4, sulfate; nh4, ammonium; oc, organic carbon; bc, black carbon; oin, other inorganic matter; awc, aerosol water content; num, aerosol number concentration; no3, nitrate; cl, chlorine; na, sodium; Esca, aerosol scattering coefficient; Eabs, aerosol absorption coefficient.

Figure 2. Schematic diagram of the binning strategy for modeling background error covariance matrix on (a) the latitude binning data or (b) the gridded data; and the vertical profiles of standard deviations ($\mu g\ kg^{-1}$) of the coarse OIN component concentration at 06:00UTC in April 2019 (c) on average over the latitude bins, (d) at Kashi city grid and (e) at the Taklimakan desert grid (i.e., 1.5 degrees east to the Kashi city).
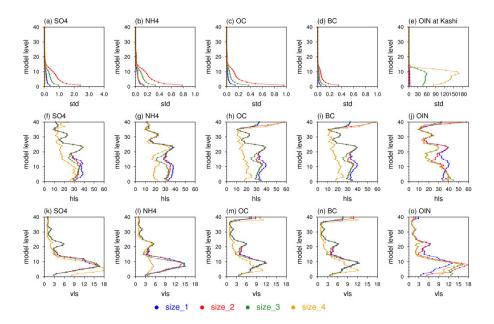
Figure 3. Background error standard deviations (std, a-e, μg kg^{-1}), horizontal correlation length scales (hls, f-j, km), and vertical correlation length scales (vls, k-o, km) at 00:00 UTC in April 2019 for the sectional sulfate, ammonium, organic aerosol (OC), black carbon (BC), and other inorganic aerosols (OIN, including dust) in the model domain 2. All the quantities in figures were the averages over the latitude bins with a half degree width, except that figure (e) represented the standard deviation of OIN at the Kashi grid.

Atmospheric
Chemistry
and Physics
Discussions

Open Access

EGU

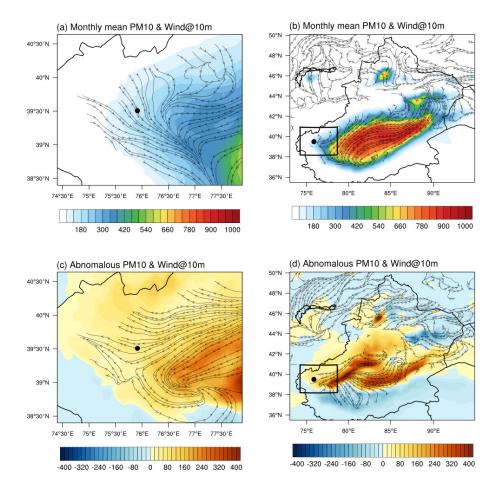Figure 4. Topography in China (a) and the model domains with the grid resolution of 20 km (b) and 5 km (c) in WRF-Chem.

Figure 5. Monthly mean (a) PM$_{10}$ concentration (µg m^{-3}) and (b) the streamlines of the 10-m wind (m s^{-1}) in April and their daily mean anomalies (c, d) during a dust storm on 24 April to the monthly mean values. Only the streamlines at the topographical height lower than 2500 meters are shown for clarity. The rectangles in figures (b) and (d) denote the fine model domain 2, which was the geographical range in the figures (a) and (c). The black points indicate the Kashi city.
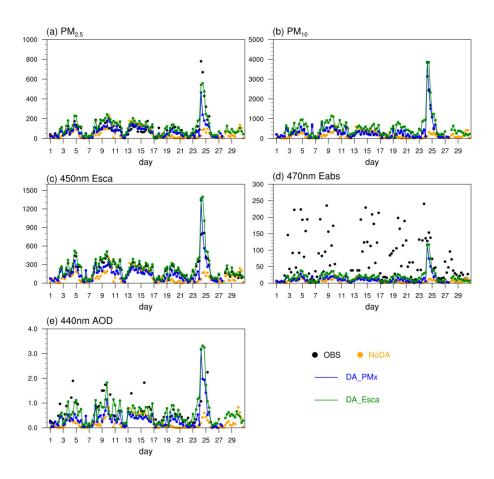
Figure 6. Comparison of (a) PM$_{2.5}$ (µg m^{-3}), (b) PM$_{10}$ (µg m^{-3}), (c) aerosol scattering coefficient (Esca, Mm^{-1}), (d) aerosol absorption coefficient (Eabs, Mm^{-1}) and (e) AOD in the observation (OBS), the background simulation (NoDA), and the DA analyses when assimilating the observed PM$_{2.5}$ and PM$_{10}$ (DA_PMx) and aerosol scattering coefficients (DA_Esca) at Kashi in April 2019.
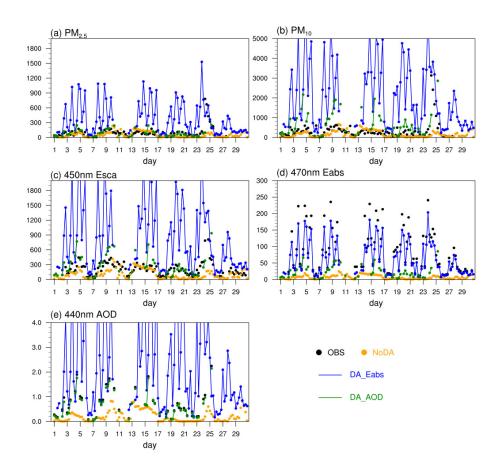
Atmospheric
Chemistry
and Physics
Discussions

Open Access

EGU

Figure 7. Comparison of (a) PM$_{2.5}$ (µg m^{-3}), (b) PM$_{10}$ (µg m^{-3}), (c) aerosol scattering coefficient (Esca, Mm^{-1}), (d) aerosol absorption coefficient (Eabs, Mm^{-1}) and (e) AOD in the observation (OBS), the background simulation (NoDA), and the DA analyses when assimilating the observed aerosol absorbing coefficients (DA_Eabs) and AOD (DA_AOD) at Kashi in April 2019.
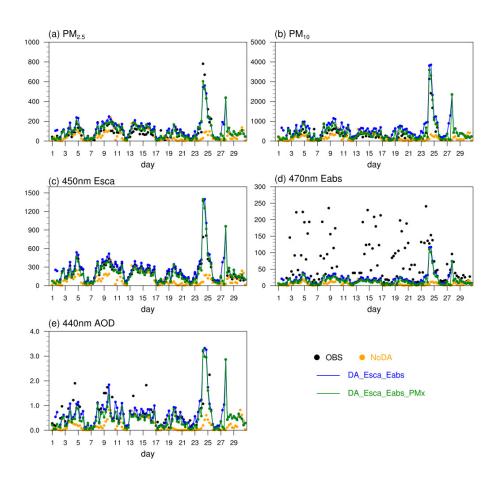
Figure 8. Comparison of (a) PM$_{2.5}$ (μg m^{-3}), (b) PM$_{10}$ (μg m^{-3}), (c) aerosol scattering coefficient (Esca, Mm^{-1}), (d) aerosol absorption coefficient (Eabs, Mm^{-1}) and (e) AOD in the observation (OBS), the background simulation (NoDA), and the DA analyses of DA_Esca_Eabs and DA_Esca_Eabs_PMx at Kashi in April 2019.
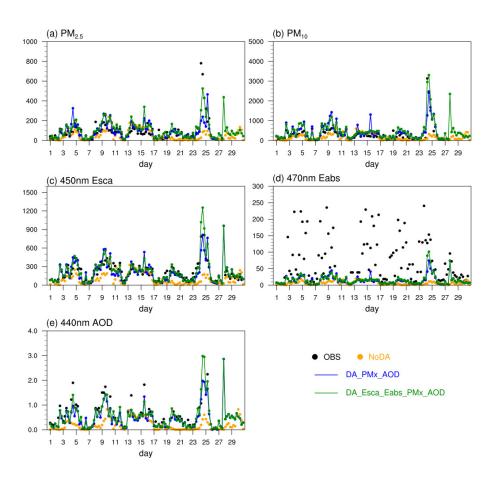
Figure 9. Comparison of (a) $PM_{2.5}$ ($\mu g\ m^{-3}$), (b) PM_{10} ($\mu g\ m^{-3}$), (c) aerosol scattering coefficient (Esca, Mm^{-1}), (d) aerosol absorption coefficient (Eabs, Mm^{-1}) and (e) AOD in the observation (OBS), the background simulation (NoDA), and the DA analyses of DA_PMx_AOD and DA_Esca_Eabs_PMx_AOD at Kashi in April 2019.
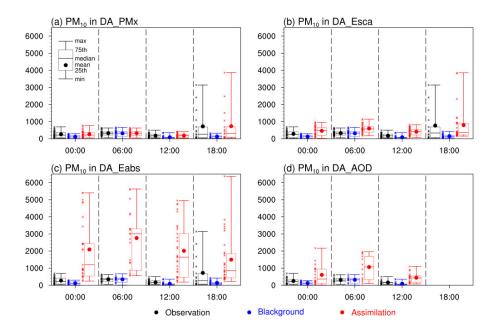
Figure 10. Surface PM_{10} concentrations (µg m^{-3}) in the observation (black), background simulation (blue) and the DA analyses (red) at 00:00, 06:00, 12:00, 18:00 UTC in April when assimilating the observations of (a) PMx, (b) aerosol scattering coefficients (Esca), (c) aerosol absorption coefficient (Eabs), and (d) AOD, respectively. The DA_AOD had no analysis at 18:00 UTC that was local midnight. Kashi is 6 hours ahead of UTC (UTC+6).
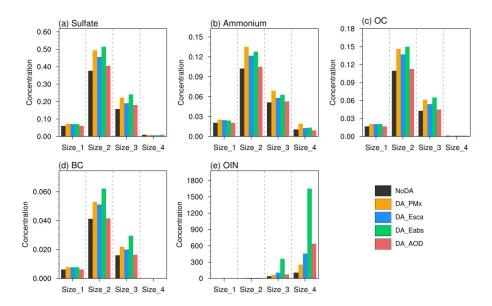
Figure 11. Monthly mean aerosol concentrations (µg m^{-3}) per size bin in the background (NoDA) and the DA experiments when assimilating the individual observation at Kashi in April 2019.
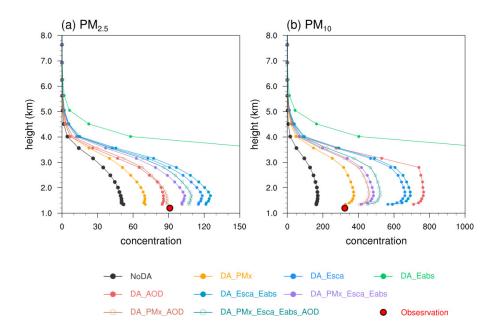
Figure 12. Monthly mean vertical concentration profiles of (a) $PM_{2.5}$ (µg m^{-3}) and (b) PM_{10} (µg m^{-3}) at Kashi in April 2019.

Figure 13. Monthly mean chemical composition in percent (%) in the PM_{10}, excluding the OIN component at Kashi in April 2019.

Table 1. The observed surface particle concentration, aerosol scattering coefficient (Esca), aerosol absorption coefficient (Eabs), and AOD used for the DA analysis and their observational errors.

	Data time range	Wavelength (nm)	Observation error (e)
PM$_{2.5}$ & PM$_{10}$ (μg m^{-3})	Apr 1 – Apr 30		$e = \sqrt{e_1^2 + e_2^2}$ $e_1 = 1.5 + 0.75 \cdot PM_x$ $e_2 = 0.5 \cdot e_1 \cdot \sqrt{\dfrac{d}{3000}}$ d: grid spacing in meter
AOD	Mar 29 – Apr 25	440, 675, 870, 1020	$e = 0.01/\text{height} \times 10^8$
Esca (Mm^{-1})	Apr 2 – Apr 30	450, 525, 635	$e = 10$
Eabs (Mm^{-1})	Apr 2 – Apr 30	470, 520, 660	$e = 10$

Table 2. The monthly mean values of the PM$_{2.5}$ and PM$_{10}$ concentrations (µg m^{-3}), 450 nm aerosol scattering coefficient (Esca, Mm^{-1}), 470 nm aerosol absorption coefficient (Eabs, Mm^{-1}) and 440 nm AOD in the background and analysis data and their correlation values (in brackets) with the observations at 00:00, 06:00, 12:00, 18:00 UTC at Kashi in April 2019. The underlined number in bold denotes the analysis in the monthly mean value that is not significantly different from the observation. The number in the bracket is the significant correlation, and the dashed line denotes an insignificant correlation. Both the statistical tests of the mean and correlation are conducted at the significance level of 0.05.

DA experiment	PM$_{2.5}$ (µg m^{-3})	PM$_{10}$ (µg m^{-3})	Esca (Mm^{-1})	Eabs (Mm^{-1})	AOD
Observation	91.0	323.2	204.5	81.9	0.70
Background	53.1 (0.23)	166.9 (-----)	106.5 (0.37)	6.7 (0.33)	0.18 (0.58)
Analysis					
DA_PMx	**70.3** (0.86)	**331.2** (0.99)	158.9 (0.86)	12.2 (0.33)	0.31 (0.37)
DA_AOD	**85.0** (0.33)	716.9 (0.35)	280.1 (0.58)	23.1 (0.16)	**0.63** (0.99)
DA_Esca	120.5 (0.93)	596.2 (0.96)	**244.6** (0.97)	19.1 (0.48)	**0.57** (0.47)
DA_Eabs	395.1 (0.31)	2l61 (0.31)	849.0 (0.65)	65.1 (0.99)	1.95 (-----)
DA_Esca_Eabs	122.9 (0.92)	604.4 (0.93)	247.7 (0.97)	20.0 (0.46)	**0.58** (0.47)
DA_PMx_Esca_Eabs	**101.1** (0.93)	**411.2** (0.98)	**198.8** (0.92)	15.2 (0.38)	0.41 (0.47)
DA_PMx_AOD	**89.5** (0.51)	**408.8** (0.75)	**195.8** (0.74)	14.9 (0.32)	**0.51** (0.95)
DA_PMx_Esca_Eabs_AOD	**107.9** (0.77)	452.9 (0.94)	**216.3** (0.92)	16.2 (0.38)	**0.53** (0.93)

Table 3. The ratios of AOD, aerosol scattering/absorption coefficient to PM_{10} concentration (mean ± standard deviation) in the observations, the model background data, and the DA analysis when assimilating surface particle concentrations (DA_PMx).

	Ratios of 440 nm AOD to PM_{10} (μg^{-1} m^3)	Ratios of 450 nm aerosol scattering coefficient to PM_{10} (Mm^{-1} μg^{-1} m^3)	Ratios of 470 nm aerosol absorption coefficient to PM_{10} (Mm^{-1} μg^{-1} m^3)
Observation	0.0033±0.0021	0.91±0.05	0.42±0.36
Background	0.0011±0.0007	0.68±0.24	0.05±0.02
Analysis	0.0012±0.0005	0.58±0.21	0.04±0.01

Table 4. The mean instantaneous clear-sky shortwave (SW), longwave (LW) and the net (SW+LW) direct radiative forcing (Wm^{-2}) at the top of atmosphere (TOA), in the atmosphere (ATM) and at the surface (SRF) in the background and the simulations restarted from the analyses of DA_PMx and DA_PMx_AOD at one hour after the analysis times of AOD DA at Kashi in April 2019.

	SW (Wm^{-2})			LW (Wm^{-2})			SW+LW (Wm^{-2})		
	TOA	ATM	SRF	TOA	ATM	SRF	TOA	ATM	SRF
Background	-7.8	+11.3	-19.1	+0.6	-3.6	+4.2	-7.2	+7.7	-14.9
DA_PMx	-8.7	+21.7	-30.4	+1.1	-7.4	+8.5	-7.6	+14.3	-21.9
DA_PMx_AOD	-12.2	+32.1	-44.3	+1.7	-12.6	+14.3	-10.5	+19.5	-30.0