Improving the Sectional MOSAIC Aerosol models of WRF-Chem with the revised Gridpoint Statistical Interpolation System and multi-wavelength aerosol optical measurements: DAO-K experiment 2019 at Kashi, near the Taklamakan Desert, northwestern China

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Abstract

The Gridpoint Statistical Interpolation data assimilation (DA) system was developed for the four-size bin sectional Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol mechanism in the Weather Research and Forecasting-Chemistry (WRF-Chem) model. The forward and adjoint operators for the aerosol optical depth (AOD) analysis were derived from WRF-Chem aerosol optical code. We applied three-dimensional variational DA to assimilate the multi-wavelength AOD, ambient aerosol scattering coefficient, and aerosol absorption coefficient, measured by the sun-sky photometer, nephelometer, and aethalometer, respectively. These were undertaken during a dust observation field campaign at Kashi in northwestern China in April 2019. The results showed that the DA analyses decreased the low biases in the model aerosols; however, it had had some deficiencies. Assimilating the surface particle concentration increased the coarse particles in the dust episodes, but AOD, and the coefficients for aerosol scattering and absorption, were still lower than observed values. Assimilating aerosol scattering coefficient separately from AOD improved the two optical quantities. However, it caused an overestimation of the particle concentrations at the surface. Assimilating the aerosol absorption coefficient yielded the highest positive bias in the surface particle concentration, aerosol scattering coefficient, and AOD. The positive biases in the DA analysis were caused by the forward operator underestimating particle scattering and absorption efficiency. As a compensation, the DA system increased particle concentrations excessively so as to fit the observed optical values. The best overall improvements were obtained from the simultaneous assimilation of the surface particle concentration and AOD. The assimilation did not substantially change the aerosol chemical fractions. After DA, the clear-sky aerosol radiative forcing at Kashi was –10.5 Wm\(^{-2}\) at the top of the atmosphere, 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.
for radiative transfer calculation. Strictly speaking, the lack of aerosol components violates the unbiased requirements for the model states in the DA system. Lack of size-segregated aerosols may introduce a bias in the calculation of optical aerosols. The official GSI can assimilate the surface particle concentration from the aerosol mechanism apart from GOCART (Zang et al., 2016), but its AOD DA is tightly bound with the GOCART aerosols. If one wished to use GSI to assimilate AOD for the other aerosol mechanisms, a compromise solution was to integrate the map of the speciated aerosols of other mechanisms into that of the GOCART aerosols. For example, Tang et al. (2017) used the official GSI to assimilate MODIS AOD with the aerosols from the Community Multi-scale Air Quality Model (CMAQ). They incorporated the map of the 54 aerosol components of CMAQ into the five CRTM aerosols and repartitioned the mass increments of each CMAQ aerosol according to the ratio of aerosol chemical components in the background field. This repartitioning is called the “ratio approach.” Cheng et al. (2019) assimilated the lidar extinction coefficient profiles measured in Beijing to modify the Weather Research and Forecasting-Chemistry (WRF-Chem) Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosols. They used the ratio approach to map eight MOSAIC aerosols based on five GOCART aerosols. This mapping strategy is readily implemented but introduces inconsistent size-segregated aerosol information (e.g., hygroscopcity and extinction efficiency) between the aerosol model and the DA system. Because building a GSI system for a new aerosol mechanism is quite technical, the official GSI for the GOCART aerosols is still a primary choice for recent aerosol DA studies (Bao et al., 2019; Xia et al., 2019; Hong et al., 2020).

Because of the shortcomings, the official GSI has been extended to cooperate with other aerosol mechanisms in WRF-Chem. The MOSAIC mechanism in WRF-Chem simulates aerosol mass and number concentrations in either four- or eight-size bins. This sectional aerosol mechanism involves nitrate chemistry and can simulate SOA with the volatility basis set scheme. Saide et al. (2013) proposed a revised GSI version that performed variational DA for the MOSAIC aerosols. The authors generated the adjoint operator code with the automatic differentiation tool (ADT), TAPENADE v3.6. The ADT used the chain rule of derivative calculus on the AOD source code in WRF-Chem. They assimilated multi-source AOD data with the MOSAIC aerosols over continental United States and found that incorporating multi-wavelength fine-mode AOD redistributed the aerosols’ particulate mass concentration sizes.

The revised GSI system assimilated Korean ground-based and geostationary satellite AOD datasets to improve local aerosol simulations (Saide et al., 2014, 2020). Pang et al. (2020) developed the official GSI to work with the Modal Aerosol Dynamics Model for Europe with the Secondary Organic Aerosol Model (MADE/SORGAM) aerosols in WRF-Chem. The authors used the WRF-Chem AOD code as the forward operator to calculate the essential aerosol optical properties, which were then inputted to the CRTM adjoint operator. Because aerosols were externally mixed in CRTM, the setting of the internal mixture per size bin in WRF-Chem was not taken into account, and the AOD of each aerosol component was calculated separately.

This study provides a solution to improve the capability of the GSI 3D-Var DA system for the sectional MOSAIC aerosols in WRF-Chem. We designed the adjoint operator code for AOD
DA based on the WRF-Chem intrinsic aerosol optical subroutine (Fast et al., 2006), that is, without using the ADT. The adjoint code is programmed based on the analytical equations of the linear tangent model for AOD. As our revised GSI does not use the CRTM module, it avoids the problem of needing to eliminate WRF-Chem aerosols characteristics (e.g., aerosol mixture state and size distribution) to meet the CRTM input requirements. The forward and adjoint operators are coordinated, since they are derived from the same WRF-Chem code, and are written in a single subroutine, which is coupled to the GSI at the place of invoking CRTM for the AOD calculation. In addition to AOD DA, our adjoint operator has two variants to assimilate the aerosol scattering and absorption coefficients, measured using a nephelometer and aethalometer, respectively.

This study verifies the effectiveness of our revised GSI system by incorporating multi-wavelength aerosol optical observations that were measured during an international field campaign, the Dust Aerosol Observation-Kashi, in April 2019 at Kashi city, neighboring the Taklamakan Desert, northwestern China. This desert is the second largest globally, and is the primary source of dust aerosols in East Asia. The dust from the desert affects the nearby Tibetan Plateau (Ge et al., 2014; Jia et al., 2015; Zhao et al., 2020), air quality and climate in East Asia (Huang et al., 2014), and the biogeochemical cycles in the western Pacific Ocean (Calil et al., 2011). A successful DA analysis will help improve the local air quality forecast and enhance our understanding of the environmental impacts of local dust storms. The remainder of this paper is organized as follows. Section 2 describes the revised GSI system, the experimental design, and the observed data. Section 3 presents the DA results when assimilating different observations. Section 4 discusses the impact of DA on aerosol chemical composition and aerosol direct radiative forcing. Finally, Section 5 provides the conclusions and limitations that need further research.

2. Methodology and Data

2.1 Forecast Model

The background aerosol fields were simulated using the WRF-Chem model version 4.0 (Grell et al., 2005; Fast et al., 2006). The model configurations included the Purdue Lin microphysics scheme (Chen and Sun, 2002), the unified Noah land surface model (Tewari et al., 2004), the Yonsei University scheme for planetary boundary layer meteorological conditions (Hong et al., 2006), and the rapid radiative transfer model for general circulation models (RRTMG) scheme for shortwave and longwave radiation (Iacono et al., 2008). The gas-phase chemistry was simulated using the carbon bond mechanism (Zaveri and Peters, 1999), including aqueous-phase chemistry. The aerosol chemistry was simulated using the MOSAIC mechanism (Zaveri et al., 2008), which simulated sulfate, nitrate, ammonium, black carbon (BC), organic carbon (OC), sodium, calcium, chloride, carbonate, and other inorganic matter (OIN, e.g., trace metals and silica). SOA was excluded from our experiments to accelerate model integration. Although ignoring that SOA biased the model, the influence was assumed to be small, based on low anthropogenic and biogenic emissions in the vicinity of the desert. We performed the MOSAIC aerosol simulations with four-size bins (0.039–0.156 μm, 0.156–0.625 μm, 0.625–2.500 μm, and 2.5–10.0 μm dry diameters). The sectional aerosol data in the hourly model output were the aerosol dry mass mixing ratios of chemical
The aerosol compositions included hydrophilic particulates (i.e., SO$_2^-$, NO$_x$, NH$_4^+$, Cl$^-$, Na$^+$) and hydrophobic particulates (i.e., BC, OC, and OIN). The dust emission was simulated using the GOCART dust scheme (Ginoux et al., 2001). The dust mass was included in the OIN concentration determination and aerosol optical calculation. The aerosol compositions were externally mixed between the size bins and internally mixed in each size bin. The internal mixing refractive index was the volume-weighted mean refractive index of each composition.

### 2.2 Assimilation System

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

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

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

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

### 2.2.1 Control Variables

The control variables in this study were the mass mixing ratio of composition of each aerosol per size bin, which corresponded to the WRF-Chem output data only. This set therefore differed from previous studies that lumped aerosols per size bin as control variables. The lumped aerosols avoided the burdensome task of specifying the background error statistics for numerous aerosols (Li et al., 2013; Pagowski et al. 2014). Although our control variables could have been further optimized, here we designed the control variable using only those that substantially contributed to the total mass concentrations. In the case of Kashi the desert, the OIN was predominant, accounting for $\sim$99% of the total particle mass concentrations. The control variable could thus have exclusively comprised the OIN.
However, because we were curious about the response of aerosol chemical fractions in the DA constraint, we set the control variables of five aerosol mass mixing ratios of SO$_4^{2-}$, NH$_4^+$, OC, BC, and OIN per size bin. Nitrate, chlorine, and sodium had miniscule background concentrations and remained the background values in the DA analysis. There were twenty control variables in total for the four-size bin simulations, and the time cost for the DA calculation for these variables was acceptable.

Our design of the control variables was different from the AOD assimilation in Saide et al. (2013), with theirs being the natural logarithm of the total mass mixing ratio per size bin, multiplied by the thickness of the model layer. As the high model layer had a significant layer thickness with low aerosol concentrations, the multiplication offset the opposite effects of increasing layer thickness versus decreasing concentrations with increase in altitude. This prevented the addition of many modifications for the high model layers, where aerosols were low in concentration. The logarithmic transformation was used to decrease the extensive value range in the control variables caused by multiplication. Since the AOD value is often smaller than one, this leads to a significant negative logarithm value and a relatively unconstrained DA system. Saide et al. (2013) introduced two weak constraints in their cost function to cut off the user-defined “extraordinarily high” and “extraordinarily low” concentrations. However, neither the logarithmic transformation, nor the multiplication using layer thickness was set in our DA system. Saide et al. (2013) repartitioned the increments of the total mass per size bin for composition of each aerosol, with the background aerosol chemical mass fractions. Our control variable was restricted to the WRF-Chem output variable, and the DA system changed the composition of each aerosol per size bin, depending on the aerosol background errors.

Consistent with the set by Pang et al. (2020), aerosol water content (AWC) was not one of the control variables in our GSI. Otherwise, the AWC might have increased contrary to the physical constraints for the loading of hydrophilic particles, and simply as a mathematical artefact. The AWC was diagnosed according to the analyzed aerosol mass concentration and the background relative humidity. The hygroscopic growth was calculated using the WRF-Chem code coupled with the revised GSI.

### 2.2.2 Adjoint Operator for PM$_x$

The adjoint operator for PM$_x$ is the gradient of the PM$_x$ concentration to the aerosol chemical mass concentration per size bin:

$$\frac{\delta [PM_x]}{\delta [C_{aer,k}]} , k = 1,..., n_{size}$$

where $n_{size}$ is the number of size bins and is equal to four in this study; [...] denotes the mass concentration (µg m$^{-3}$ for PM$_x$); $C_{aer,k}$ is the aerosol mass mixing ratio (µg kg$^{-1}$) of SO$_4^{2-}$, NH$_4^+$, OC, BC, and OIN at the $k$-th size bin. The threshold of aerosol mass mixing ratio that yields the non-zero adjoint operator is 0.01 µg kg$^{-1}$. The PM$_{2.5}$ and PM$_{10}$ are assimilated in the
same way. When the fine and coarse particles are assimilated simultaneously, we assimilate
the concentration of PM$_{2.5}$ and the coarse particulate (PM$_{10}$-PM$_{2.5}$).

2.2.3 Forward Operator for AOD in WRF-Chem

We used the original forward operator in WRF-Chem for the aerosol optical parameters (Fast
et al., 2006). AOD is calculated as a function of wavelength according to Mie theory. The
columnar AOD $\tau$ is the sum of layer AOD across the $n_e$ model layers:

$$\tau = \sum_{z=1}^{n_z} r_z = \sum_{z=1}^{n_z} \sum_{k=1}^{n_{size}} e_{ext,z,k} \cdot n_{z,k} \cdot H_z$$  \hspace{1cm} (3)

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

$$e_{ext,z,k} = p_{ext,z,k} \cdot \pi \cdot r_{wet,z,k}^2$$  \hspace{1cm} (4)

where $p_{ext,z,k}$ is the extinction coefficient, given the desired mixing refractive indexes and the
wet particle radius. The $p_{ext,z,k}$ is attained through the Chebyshev polynomial interpolation:

$$p_{ext,z,k} = \exp \left\{ \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot e_{ext,z,k}(j) \right\}$$  \hspace{1cm} (5)

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

$$n_{z,k} = \sum_{i} \frac{m_{i,z,k}}{\rho_i} \cdot \frac{3}{4\pi \cdot r_{wet,z,k}^2} = \sum_{i} \frac{m_{i,z,k}}{\rho_i} \cdot \frac{3}{4\pi \cdot r_{dry,z,k}^3}$$ \hspace{1cm} (6)

2.2.4 Adjoint Operator Developed for AOD

As per the forward operator in Eq. (3) in WRF-Chem, we developed the adjoint operator for
AOD, which requires the derivative of $\tau$ in Eq. (3) to the mass concentration, $m_{i,z,k}$:
\[
\frac{\delta \tau}{\delta m_{i,z,k}} = \frac{\delta \tau_z}{\delta m_{i,z,k}} + \frac{\delta e_{ext,z,k} \cdot n_{z,k} \cdot H_z}{\delta m_{i,z,k}} + \frac{\delta e_{ext,z,k} \cdot \delta n_{z,k} \cdot H_z}{\delta m_{i,z,k}} + \frac{\delta e_{ext,z,k} \cdot n_{z,k} \cdot \delta H_z}{\delta m_{i,z,k}}
\]

(7)

The first term on the righthand side of Eq. (7) indicates the change in AOD as the perturbation of extinction cross section. According to Eq. (4), considering that the particle radius is constant, \( \delta e_{ext,z,k} \) is represented as:

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

(8)

where \( \delta c(h) = 0 \) assuming that the particle radius is constant. Equation (8) is expanded with the derivative of Eq. (5):

\[
\delta p_{ext,z,k} = p_{ext,z,k} \cdot \left\{ \sum_{j=1}^{n_{coef}} c_{ch}(j) \cdot \delta c_{ext,z,k}(j) \right\}
\]

(9)

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

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

(10)

where the four parameters of \( E_{ext} \) indicate the extinction efficiencies in the Mie table surrounding the point with the desired mixing refractive indexes, and the wet particle radius. The changes in interpolation weights \( \delta w \) are determined as:

\[
\begin{align*}
\delta w_{00} &= (v - 1) \delta u + (u - 1) \delta v \\
\delta w_{01} &= (1 - v) \delta u - u \delta v \\
\delta w_{10} &= (1 - u) \delta v - v \delta u \\
\delta w_{11} &= u \delta v + v \delta u
\end{align*}
\]

(11)

where

\[
\begin{align*}
\delta u &= \frac{R_{mix} - R_{low}}{R_{up} - R_{low}} \\
\delta v &= \frac{I_{mix} - I_{low}}{I_{up} - I_{low}}
\end{align*}
\]

(12)

In Eq. (12), \( R_{mix} \) and \( I_{mix} \) are the aerosol volume-weighted mean real and imaginary refractive indices, respectively. \( R_{up} \) (\( I_{up} \)) and \( R_{low} \) (\( I_{low} \)) are the nearest upper and lower limits for \( R \) (\( I \)) in the Mie table. Considering \( V_{wet,z,k} \) is the volume of all aerosol masses, the real and imaginary indices and their derivatives are:
\[ R_{\text{mix},x,k} = \sum_i n_{\text{wet, aer}} R_i \cdot \frac{m_{i,x,k}}{\rho_i \cdot V_{\text{wet},x,k}} \]
\[ \delta R_{\text{mix},x,k} = \frac{R_i}{\rho_i \cdot V_{\text{wet},x,k}} \cdot \delta m_{i,x,k} \]
\[ I_{\text{mix},x,k} = \sum_i n_{\text{wet, aer}} I_i \cdot \frac{m_{i,x,k}}{\rho_i \cdot V_{\text{wet},x,k}} \]
\[ \delta I_{\text{mix},x,k} = \frac{I_i}{\rho_i \cdot V_{\text{wet},x,k}} \cdot \delta m_{i,x,k} \]

where
\[ V_{\text{wet},x,k} = \sum_i \frac{m_{i,x,k}}{\rho_i} \]

(13)

Put Eq. (10), Eq. (11) into Eq. (9) leads to:
\[ \delta p_{z,k} = [(u - 1)\alpha_{00} + (1 - v)\alpha_{01} - v\alpha_{10} + v\alpha_{11}]\delta u + [(u - 1)\alpha_{00} - u\alpha_{01} + (1 - u)\alpha_{10} + u\alpha_{11}]\delta v \]

(15)

where
\[ \alpha_{00} = p_{\text{ext},x,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{\text{ext,00}}(j) \]
\[ \alpha_{01} = p_{\text{ext},x,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{\text{ext,01}}(j) \]
\[ \alpha_{10} = p_{\text{ext},x,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{\text{ext,10}}(j) \]
\[ \alpha_{11} = p_{\text{ext},x,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{\text{ext,11}}(j) \]

(16)

The first term on the righthand side of Eq. (7) is determined using Eq. (8) and Eq. (15). The second term on the righthand side of Eq. (7) indicates the linkage of the aerosol number and mass concentrations. It is the derivative of Eq. (6) by assuming a constant radius:
\[ \delta n_{z,k} = \frac{3 \cdot \delta m_{i,x,k}}{4\pi \cdot r_{3\text{yr},z,k}^3 \cdot \rho_i} \]

(17)

The third term on the righthand side of Eq. (7) contains the derivative of the layer thickness to the concentrations in this layer. This indicates that the light attenuation length based on per unit concentration, which can be intuitively represented by the ratio of layer thickness to the aerosol mass concentration in this layer. Putting Eq. (8) and Eq. (17) into Eq. (7), we have the original formula of the adjoint operator for AOD for the aerosol mass concentration:
\[
\frac{\delta \tau}{\delta m_{i,k}} = \frac{\delta e_{ext,z,k} \cdot n_{z,k} \cdot H_z}{\delta m_{i,k}} + \frac{e_{ext,z,k} \cdot \delta n_{z,k} \cdot H_z}{\delta m_{i,k}} + e_{ext,z,k} \cdot \frac{\delta H_z}{\delta m_{i,k}} = \\
\frac{\{[(v - 1)\alpha_0 + (1 - v)\alpha_{10} - v\alpha_{10} + v\alpha_{11}] \cdot \frac{\pi \cdot r_{wet,z,k}^2 \cdot R_i \cdot n_{z,k} \cdot H_z}{\rho_i \cdot V_{z,k} \cdot (R_{up,k} - R_{low,k})} + }{3e_{ext,z,k} \cdot \frac{H_z}{m_{i,k}}} \cdot \beta 
\]

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

\[
\frac{\delta (e_{ext,z,k} \cdot n_{z,k})}{\delta m_{i,k}} = \frac{H_z}{\sum H_z} \cdot \frac{\delta e_{ext,z,k} \cdot n_{z,k}}{\delta m_{i,k}} = \frac{H_z}{\sum H_z} \cdot \left[ \frac{e_{ext,z,k} \cdot \delta n_{z,k}}{\delta m_{i,k}} + \frac{e_{ext,z,k} \cdot \delta H_z}{\delta m_{i,k}} \right] = \\
\frac{\{[(v - 1)\alpha_0 + (1 - v)\alpha_{10} - v\alpha_{10} + v\alpha_{11}] \cdot \frac{\pi \cdot r_{wet,z,k}^2 \cdot R_i \cdot n_{z,k}}{\rho_i \cdot V_{z,k} \cdot (R_{up,k} - R_{low,k})} + }{3e_{ext,z,k} \cdot \frac{H_z}{m_{i,k}}} \cdot \beta \cdot \frac{H_z}{\sum H_z} 
\]

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

### 2.2.5 Adjoint Operator Developed for Surface Aerosol Attenuation Coefficients

The aerosol scattering and absorption coefficients measured by the nephelometer and aethalometer, respectively, are similar to the aerosol extinction coefficient at the surface in Eq. (19). Neither of the two coefficients address the layer thickness. The adjoint operator for the aerosol scattering coefficient measured by nephelometer is described as follows:
\[
\delta(e_{sca,1,k} \cdot n_{1,k}) \over \delta m_{l,1,k} = \left\{ \left[ (u - 1) \alpha_{00} + (1 - u) \alpha_{01} - u \alpha_{10} + u \alpha_{11} \right] \cdot \pi \cdot r_{\text{net,1,k}}^2 \cdot R_i \cdot n_{1,k} \right. \\
\left. + \frac{3 e_{sca,1,k}}{4 \pi \cdot r_{\text{dry,1,k}}^3 \cdot \rho_l} \right\} \cdot \beta
\]

(20)

\[
a_{00} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{sca,00}(j) \quad a_{01} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{sca,01}(j)
\]

\[
a_{10} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{sca,10}(j) \quad a_{11} = p_{sca,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{sca,11}(j)
\]

(21)

The adjoint operator for the aerosol absorption coefficient measured by aethalometer is

\[
\delta(e_{abs,1,k} \cdot n_{1,k}) \over \delta m_{l,1,k} = \left\{ \left[ (u - 1) \alpha_{00} - u \alpha_{01} + (1 - u) \alpha_{10} + u \alpha_{11} \right] \cdot \pi \cdot r_{\text{net,1,k}}^2 \cdot I_j \cdot n_{1,k} \right. \\
\left. + \frac{3 e_{abs,1,k}}{4 \pi \cdot r_{\text{dry,1,k}}^3 \cdot \rho_l} \right\} \cdot \beta
\]

(22)

\[
a_{00} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{abs,00}(j) \quad a_{01} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{abs,01}(j)
\]

\[
a_{10} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{abs,10}(j) \quad a_{11} = p_{abs,1,k} \cdot \sum_{j=1}^{n_{\text{coef}}} c_{ch}(j) \cdot E_{abs,11}(j)
\]

(23)

where the symbols have the same meaning as before. The subscript one denotes the surface layer, while \(sca\) and \(abs\) denote “scattering” and “absorption,” respectively.

As shown in the adjoint operators, the gradients of the aerosol mass concentrations rely on the aerosol number concentration; meanwhile, the number concentration is estimated according to the mass concentration and the particle radius. The two concentrations are intertwined in the DA system, indicating the nonlinearity of the adjoint operator. This nonlinearity is handled with a succeeding minimization of the cost function within the GSI. That is, the cost function is first minimized with the number concentration in the background field, and the number concentration is updated with the first analyzed aerosol mass concentrations. In the second minimization, the number concentration assessed in the first analysis constructs a new adjoint.
opera value, resulting in a new analysis of mass concentrations. This iterative process is
denoted as the “outer loop,” which is repeated several times to attain the final analysis
(Massart et al., 2010). We used ten iterations to handle the nonlinearity in the adjoint
operator. The WRF-Chem AOD code is coupled into the GSI subroutine at the place of
invoking CRTM. The AOD and the adjoint operators of Eq. (19), Eq. (20), and Eq. (22) are
simultaneously determined in a single subroutine, which is cyclically invoked in the outer
loops within the GSI.

2.3 Background Error Covariance (BEC)

Many aerosol DA studies used the National Meteorological Center (NMC) method (Parrish
and Derber, 1992) to model the BEC matrix. The NMC method uses long-term archived
weather data that are created in the forecast cycles. It computes the statistical differences
between two forecasts with different leading lengths (e.g., 24 h and 48 h), but which are valid
at the same time. The NMC method is workable because solving global weather forecasts is
an initial value problem of mathematical physics. That is, a slight difference in the initial
atmospheric state would lead to a substantially different prediction, because of the chaos in
the atmosphere. However, a regional model is a boundary value problem. Meteorological
reanalysis data drive the regional chemistry simulation, and the driving data quality affects the
simulation (Giorgi and Mearns, 1999). The WRF-Chem simulations in the NMC method only
reflected the influences of using different initial conditions. As the model runs, the influence
of the initial conditions becomes weak, while the influence of lateral boundary conditions
always takes effect. Because the same reanalysis data drive the paring regional model
simulations, the following lateral boundary conditions for the simulations of the two leading-
lengths are similar. This leads to a limited regional model difference when using the NMC
method. We speculate that the NMC method cannot fully represent the model biases in
emission inventories and model chemistry, and it underestimates the aerosol error in WRF-
Chem.

Some aerosol DA studies have created background error variance using the ensemble
simulations by randomly disturbing model lateral boundary conditions and surface emissions
(Peng et al., 2017; Ma et al., 2020). The ensemble experiments better represent the model
error, but significantly increase the computational burden. Here, we used the standard
deviation of hourly aerosol concentrations in April in the background field (first guess field)
to represent the background error variance. The rationale of this approach is that the Tarim
Basin acts as a “dust reservoir” and traps dust particles for a period, before being carried long-
distance by wind (Fan et al., 2020). The model bias in dust dominates the model aerosol error,
and is correlated with the aerosol variation as the weather fluctuates. The model bias is small
on clear days when the aerosol concentration is low. Conversely, the bias is large when the
mean concentration is high: that is, on heavily-polluted days. Because the mean aerosol
concentration correlated positively with the aerosol variation, we used the standard deviation
of aerosol concentration to represent the background aerosol error. This approach was similar
to Sic et al. (2016), who set a percentage of the first guess field for the background error
variance. Our approach prioritizes DA modification of aerosols which have high background
mean concentrations.
We calculated the statistics of the background error, including the aerosol standard deviation and the horizontal and vertical correlation length scales, using the GENerate the Background Errors (GEN-BE) software (Descombes et al., 2015), based on the one-month hourly aerosol concentrations in WRF-Chem. We obtained the statistics of four static BECs for the four DA analysis hours (i.e., 00:00, 06:00, 12:00, and 18:00 UTC), respectively. The DA procedures for the April 2019 data repeatedly use the statistics of the background error at the corresponding analysis time. A usual strategy to enrich the samples of model results for calculating the statistics is to gather model grid points with similar characteristics of the atmosphere, referred to as “binning.” The statistics are spatially averaged over the binned grid points. The default strategy in the GEN_BE for GSI is latitude-binning, which creates a latitude-dependent error correlation function (Figure 2a). The latitude binning is generally used for latitude flow dependency and works for large and global domains (Wu et al., 2002).

However, we found that using the latitude-binning strategy overestimated the surface PM$_{10}$ concentration when assimilating aerosol optical observations. One reason for this was related to the model bias in particle extinction efficiency, as discussed in Section 3.3. Another plausible reason is related to the vertical profile of the background model error. The maximum dust error occurred at the surface of the desert (Figure 2e) because of the local dust emission sources, but the maximum error at Kashi was at the dust transporting layer above the surface (Figure 2d). Owing to the vast extent of the Taklamakan Desert, the latitude-binning suppressed the local error characteristics at Kashi, and led to a vertical error profile (Figure 2c) similar to that over the desert (Figure 2e).

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

The horizontal and vertical correlation length scales determine the range of observation innovations spreading from the observation locations. The horizontal influences had small changes in altitude within the lowest 15 model layers (below a height of ~5 km) (Figures 3f-j), indicating that the dust transport layer was well-mixed in the lower atmosphere. This deep dust layer was consistent with the dust simulation by Meng et al. (2019). They showed that the dust in spring was vertically mixed in a thick boundary layer to a height of 3–5 km in the Tarim Basin. The vertical correlation length scales first increased from low values at the surface, to high values at ~2.5 km in height (for the 8–9 layers), indicating that strong winds...
yielded intense aerosol upward flux. The vertical correlation length scale quickly decreased from the maximum value, with further increase in altitude corresponding to the large particle gradient at the upper edge of the transporting layer. The latter was associated with laminar air motion during the dust storm.

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

### 2.4 Observational Data and Errors

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

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

### 2.5 Experimental Design

The WRF-Chem simulations were configured in a two-nested domain centered at 82.9°E, 41.5°N. The coarse domain was a 120×100 (west-east × north-south) grid with a horizontal resolution of 20 km that covered the Taklamakan Desert, and the fine domain was an 81×61 grid with a resolution of 5 km, focusing on Kashi and environs (Figure 4a). Both domains had 41 vertical levels extending from the surface to 50 hPa. The two domains were two-way coupled. The parent domain covered the entire dust emission source, providing dust transport fluxes at the lateral boundaries of the fine domain. The aerosol radiative effect was set to provide feedback on the meteorology. The indirect effect of aerosols was not set in the
experiments. Initial and lateral boundary meteorological conditions for WRF-Chem were the one-degree resolution of the National Centers for Environmental Prediction Final Analysis data created by the Global Forecast System model. The meteorological lateral boundary conditions for the coarse domain were updated every six hours, and were linearly interpolated between the updates in WRF-Chem. We did not set the chemical boundary conditions for the coarse domain. The Multiresolution Emission Inventory of China (MEIC) for 2010 (www.meicmodel.org) provided anthropogenic emission levels. The biogenic emission levels were estimated online using the Model of Emissions of Gases and Aerosols from Nature (Guenther et al., 2006). Wildfire emissions were not set in the experiments.

We conducted a one-month WRF-Chem simulation for April 2019, starting at 00:00 UTC 27 March and discarding the first five days for spin-up. The revised GSI system modified the aerosols in the fine domain at 00:00, 06:00, 12:00, and 18:00 UTC on each day, starting from 00:00 UTC 1 April until the end of the month. We assimilated the observations four times a day because the reanalyzed meteorological data were available for the four time slices, which facilitated the model restarting from the DA analyses. The hourly PM\(_s\) observations were assimilated at the exact time of analysis. The observed AOD and aerosol scattering/absorption coefficients were assimilated when they fell within ±3 h, centered at the time of analysis. Table 2 shows the DA experiments. The literal meanings of the experimental names denote the observations that were individually or simultaneously assimilated. To study the impact of DA on aerosol direct radiative forcing (ADRF), we restarted the WRF-Chem model from each DA analysis, which then ran to the next analysis time. Each running performed the radiation transfer calculation both with, and without aerosols, respectively. The irradiance difference between the two pairing calls was aerosol radiative forcing. Section 4.2 shows the DA effects on the clear-sky ADRF values.

| Table 2, Figure 4 |

3. Results

3.1 Evaluation of Control Experiment

Table 2 shows the monthly mean values and correlations between the observed data and the model results. The statistical values were based on the pairing data between the model results and the observations. Figures 6–9 show the surface PM\(_s\) concentrations, aerosol scattering, absorption coefficients, and AOD when assimilating the observations at 00:00, 06:00, 12:00, and 18:00 UTC each day in April.

Kashi is in the junction between the Tian Shan Mountains to the west and the Taklamakan Desert to the east (Figure 5a). In the Tarim Basin, the prevailing surface wind is easterly or northeasterly, which raises dust levels and carries the particles westward (Figure 5b). An intense dust storm hit the city at noon on 24 April 2019, with a peak PM\(_{10}\) concentration exceeding 3,000 μg m\(^{-3}\). The dust storm travelled across the northern part of the desert and carried the dust particles to Kashi and the mountainous area (Figure 5c, d). A few mild dust storms occurred at Kashi on April 3–5, April 8–11, and April 14–17 (Figure 6b), and the maximum PM\(_{10}\) concentrations were in the range of 400–600 μgm\(^{-3}\). The time series of PM\(_{2.5}\),
aerosol scattering/absorption coefficient, and AOD showed patterns, similar to those for PM$_{10}$ (Figure 6).

WRF-Chem captured the main dust episodes, but significantly underestimated the aerosols at Kashi (Table 2). The background monthly mean concentrations of PM$_{2.5}$ and PM$_{10}$ were half of the observed values, with a low correlation ($R < 0.3$). The simulated dust storm on 24 April was a mild dust event and had a maximum PM$_{10}$ of ~300 µg m$^{-3}$, one-tenth of the observed value. The model lowered the aerosol scattering/absorption coefficients and AOD by 50–90%.

The OIN component accounted for the model bias in PM$_{10}$ on dusty days. Zhao et al. (2020) proposed that the GOCART scheme reproduced dust emission fluxes under conditions of weak wind erosion but underestimated the emissions in conditions of strong wind erosion. We did not assimilate meteorology. The model bias in the surface wind introduces errors in dust emission, and places bias on the number of dust particles entering the city. In the non-dust days, for example, on 20 April, the model hourly PM$_{2.5}$ was 10–50% of the observed data levels. The simulated anthropogenic aerosols were probably too low to be reasonable for this city. The residential sector is a major source of anthropogenic emissions, including PM$_{2.5}$, BC, and OC, particularly in the developing western area. The latter sector accounts for 36–82% of these emissions, according to the MEIC emission inventory (Li et al., 2017). The sector is the primary source of uncertainty in anthropogenic emissions inventories in China. We speculated that the low bias in anthropogenic emissions could be significant for Kashi, resulting in low anthropogenic aerosols in the model.

3.2 Assimilating PM$_{2.5}$ and PM$_{10}$ Concentrations
Simultaneous assimilation of the observed PM$_x$ (DA_PM$_x$) improved both the fine and coarse particle concentrations, with a substantial improvement in the third and fourth particle sizes of the OIN composition (Figure 11e). The analyzed monthly mean PM$_{10}$ increased to 331.2 µg m$^{-3}$, with a high correlation of 0.99. The analyzed monthly mean PM$_{2.5}$ was increased to 70.3 µg m$^{-3}$, although it was still lower than the observed levels, with a high correlation of 0.86. The low bias in PM$_{2.5}$ was mainly in the dust storm on 24–25 April (Figure 6a). This indicates that the DA system preferentially modified the coarse particle concentrations because the coarse particles were assigned with a high background model error according to our BEC modeling strategy. Intentionally, this modification that mainly focused on the highest concentration of coarse particles was reasonable. It decreased the model biases by raising the heaviest loading aerosols. As the particle concentration increased, the aerosol scattering coefficient increased to 158.9 Mm$^{-1}$, with a high correlation of 0.86. However, the improvements in the analyzed absorption coefficients and AOD were insufficient (Figures 6d-e). The analyzed absorption coefficient was 12.2 Mm$^{-1}$, 85% lower than observed levels, with a low correlation of 0.33. The analyzed AOD showed a monthly mean value of 0.31, 56% lower than observed levels, with a low correlation of 0.37.
Figure 10 shows the diurnal concentrations of PM$_{10}$ in the analyses in April. The observed PM$_{10}$ showed a substantial variation at 18:00 UTC, the (local midnight). This substantial nocturnal variation was partly owing to the dust storm that started on 24 April and ended the next day. This midnight variation was also related to a nocturnal low-level jet. Ge et al. (2016) pointed out that there was a nocturnal low-level jet at a height of 100–400 m, with a wind speed of 4–10 m s$^{-1}$ throughout the year in the Tarim Basin. They stressed that the low-level jet broke down in the morning, transporting its momentum toward the surface, and increased dust emissions. The nocturnal low-level jet increased the possibility of dust particles moving towards the city at night, causing a high PM$_{10}$ variation at 18:00 UTC. The diurnal changes in the DA analyses followed the observed levels, but had higher mean values (Figure 10a).

3.3 Assimilating AOD

Assimilating AOD (DA_AOD) improved the monthly mean AOD to 0.63, with a high correlation of 0.99 (Figure 7e). The monthly mean PM$_{2.5}$ was improved to 85.0 µg m$^{-3}$, quite close to the observed level, but the analyzed PM$_{10}$ was 716.9 µg m$^{-3}$, which was more than double the observed value. The DA system improved the AOD at the price of deteriorating the data quality of surface particle concentrations, opposite to the result when assimilating PM$_x$. Surface particle overestimations have been reported in previous studies (Liu et al., 2011; Ma et al., 2020; Saide et al., 2020). Ma et al. (2020) assimilated ground-based lidars and PM$_{2.5}$ simultaneously in eastern China using the WRF-Chem/DART (Data Assimilation Research Testbed). They claimed that WRF-Chem underestimated the AOD and low-level aerosol extinction coefficient because the model had a low bias in relative humidity, which led to less AWC and lowered the single-particle extinction. As a compensation, the DA system overestimated the total particle concentration to fit the observed AOD value. In the arid area of Kashi, PM$_{10}$ was strongly overestimated when assimilating AOD. We speculate that WRF-Chem also lowers the dust extinction efficiency.

Table 3 shows the ratios of the AOD and aerosol scattering/absorption coefficients to the surface PM$_{10}$ concentrations in the DA_PM$_x$ experiment. The other DA experiments yielded almost similar results. This shows that the ratio of AOD to PM$_{10}$ in the background model result was only one-third of the observed levels. This low ratio indicated a model low bias in particle scattering/absorption efficiency and imposed the DA system to overestimate the PM$_{10}$ to fit the observed AOD data. The low bias is related to the aerosol optical module, which is based on Mie theory in WRF-Chem. First, the simulations used four-size bin particle segregation. This coarse size representation aggregated many aerosols in the accumulation mode. Because small particles have a strong of light attenuation capability, according to the Mie theory, too many coarse particles would not effectively increase the AOD. Saide et al. (2020) linked the aerosol optics to the size bin representation (from 4 to 16 bins) for hazes in South Korea. They showed that WRF-Chem underestimated the dry aerosol extinction, and the underestimation could be relieved when using a finer size bin than four. Okada and Kai (2004) found that the dust particle radius in the Taklamakan Desert was in the range of 0.1–4 µm, indicating the dominant fine-mode particles in the desert. Using the four-size bin would simultaneously obtain better analyses of both AOD and PM$_x$. Second, the dust particles are
irregular in shape (Okada and Kai, 2004), while the spherical particle is a common assumption for the aerosol optics in the Mie theory in current models, which is an essential source of uncertainty in the forward operator of WRF-Chem when the assumption of spherical particles for dust fails.

Another reason for the low ratio of AOD to PM10 is related to our approach for modeling BEC. It is important to remember that our BEC represents the possible error effects owing to model bias in aerosols. The coarse particle accounts for a large mass portion of PM10, and its bias dominates the model error. However, we cannot say that this background error assessment is unbiased. As our BEC gave a high background error to the coarse particle for its sufficient concentration, the DA system tended to increase PM10, which was not as effective in increasing AOD as PM2.5. If the background error of the coarse particle were too high, the BEC would falsely lower the ratio of AOD to PM10 in the analysis.

To reduce the overestimate in PM10 concentrations, we set the gridded standard deviation for the OIN for Kashi in place of the latitude-binning standard deviation, as discussed in Section 2.3. Figure 12 shows the analyzed vertical profiles of PM10 concentrations. Higher concentrations were observed in the low atmosphere than at the surface. These vertical error profiles decreased the surface particles and tended to increase the ratio, contrary to the effects of low model bias in particle extinction efficiency and the possible high bias in the BEC values of coarse particles. For the net effect of the compensation, the ratio in the analysis was still almost equivalent to the background value (Table 3). That is, our tuned BEC vertical profile at Kashi, to some extent canceled out the effects of other model error sources (e.g., the positive bias in the coarse particle of BEC, and the low bias in extinction efficiency) but was not sufficient to increase the ratio to the observed value. Finer aerosol size representation and a better advanced aerosol optical calculation for dust are essential solutions.

Because the DA system overestimated the aerosol number concentration, resulting in a positive bias in PM10, the analyzed aerosol scattering coefficient was overestimated up to 280.1 Mm$^{-1}$, 37% higher than the observed value. In contrast, the analyzed absorption coefficient was 23.1 Mm$^{-1}$, 72% lower than the observed value. This indicates that WRF-Chem strongly underestimated the single-particle absorption efficiency, and the low bias was too strong to be compensated by the overestimated aerosol number concentration.

Assimilating the AOD increased the diurnal variation in the DA analyses. There was a higher increase in the concentration at noon (06:00 UTC) (Figure 10d). At the hot time of the day, intense sunlight increased the light extinction by the particles. The DA system had to raise the PM10 to fit the observed high AOD values. At dawn (00:00 UTC) or dusk (12:00 UTC), when the sunlight was weak, the DA modifications were small, and the DA increases in the PM10 fell to low levels. However, because the AOD constraint was only available in the daytime and the AOD DA data were not always available as the data quality control (i.e., cloud screening), assimilating AOD did not substantially increase the correlation of PM10. The analyzed PM10 in the DA experiments still had low correlations with the observed levels ($R=0.33$–0.35).
3.4 Assimilating Aerosol Scattering Coefficient

Assimilating the aerosol scattering coefficient (DA_Esca) yielded overall analyses similar to the phenomenon in DA_AOD. The aerosol scattering coefficient was reasonably good in the analysis, with a monthly mean value of 244.6 Mm⁻¹ and a high correlation of 0.97. The analyzed monthly mean AOD was 0.57, better than the AOD of 0.31 when assimilating PM₁₀. However, the surface particle concentrations were overestimated (i.e., positive biases by 32%) for PM₂.₅, and 84% for PM₁₀, with a substantial increase in the coarse particle of OIN. Overestimations appeared during the mild dust episodes (Figure 6b). This again indicated that WRF-Chem underestimated the particle scattering efficiency, which was represented by the ratio of the scattering coefficient to PM₁₀ (Table 3). The DA system thus overfitted the PM₈ concentration to approach the observed scattering coefficient. The diurnal PM₁₀ in the analysis was similar to the assimilation of PM₈, showing a maximum improvement and a robust nocturnal variation at 18:00 UTC. Assimilating the scattering coefficient failed to improve the absorption coefficient. The monthly mean absorption coefficient was 19.1 Mm⁻¹, 77% lower than the observed value.

3.5 Assimilating Aerosol Absorption Coefficient

In contrast to the above results, assimilating the absorption coefficient (DA_Eabs) degraded all the analyses other than the absorption coefficient itself. The analyses showed substantial daily variations, and strong positive biases appeared in the dust episodes (Figure 7). The PM₂.₅ was overestimated by a factor of four, and the PM₁₀ was overestimated by a factor of six. The increases occurred each hour and enlarged the diurnal variation of PM₁₀ (Figure 10c). The maximum increase in the mean value was at 06:00 UTC, also because of the strong noontime heating in the model. As the particle concentration increased, the aerosol scattering coefficient was overfitted to 849.0 Mm⁻¹, higher than the observed levels by a factor of four. The monthly mean AOD was improbably up to 1.95. The improvement of the absorption coefficient (which was 65.1 Mm⁻¹) was insufficient, and was 21% lower than the observed levels.

Unlike DA_AOD and DA_Esca, assimilating the absorption coefficient cannot increase the absorption data at the cost of PM₁₀ overestimation. This DA failure in assimilating the absorption coefficient indicates the model biases in the representation of the particle mixture and the other aborting particles (e.g., brown carbon and aged dust). With respect to the current model, this failure is related to the aerosol absorption represented in WRF-Chem. The leading absorption aerosol in WRF-Chem is BC. This BC particle in the second size (0.156–0.625 μm) had the maximum absorption, according to Mie theory, and had the maximum DA modifications in the second-size bin (Figure 11d). However, because the BC had a small background concentration, the BC showed a small DA improvement and had small effects on increasing the particle absorption. Meanwhile, the coarse dust particle concentration was primarily increased, but the dust particles did not have a strong absorption as BC. As a result, the model lowered the ratio of the absorption coefficient of PM₁₀ by an order of magnitude (Table 3). To fit the observed absorption coefficient, the DA system dramatically...
overestimated the particle concentrations, aerosol scattering coefficient, and AOD, but the analyzed absorption coefficient was still underestimated.

3.6 Assimilating Multi-source Observations

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

In our case, simultaneous assimilation of the scattering and absorption coefficients (DA_Esca_Eabs) resulted in the analyses when assimilating the scattering coefficient alone (DA_Esca), and the inferior analysis in DA_Eabs vanished. This was because incorporating the scattering coefficient constrained the aerosol number concentrations, which also benefited from incorporating the observed absorption coefficient. Compared with the analysis assimilating the PM$_{10}$ alone (DA_PM$_{10}$), assimilating the two aerosol attenuation coefficients (DA_Esca_Eabs) better reproduced the AOD, but overestimated the surface particle concentrations. In Figures 8–9, there were extremely high values on 28 April 2019, because the scattering coefficient was missing at that time, during which the DA system assimilated the absorption coefficient alone and worsened the analysis again. Simultaneous assimilation of the surface particle concentration and the two aerosol attenuation coefficients (DA_PM$_{10}$_Esca_Eabs) improved these three assimilated parameters, but still gave a notable low bias in AOD, 41% lower than the observed levels. Simultaneous assimilation of PM$_{10}$ and AOD (DA_PM$_{10}$_AOD) gave the best overall DA results, in which all the analyses except the absorption coefficient were not significantly different in the month mean values from the observations. The analyses between DA_PM$_{10}$_AOD and DA_PM$_{10}$_Esca_Eabs were comparable, except that the former additionally increased AOD better. Simultaneous assimilation of all observations (DA_PM$_{10}$_Esca_Eabs_AOD) did not substantially improve the analyses when compared with DA_PM$_{10}$_AOD because the surface coefficients, and AOD had overlapped information of the light attenuation. A redundant information source did not introduce extra constraints on the DA system. All the DA experiments failed to improve the aerosol absorption coefficient, which always showed strong, low biases (> 76%) and low correlations (< 0.5) in the analyses, implying room for improvement of our DA system.

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

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

4. Discussion

4.1 DA Impact on Aerosol Chemical Composition

The maximum concentrations of sulfate, ammonium, BC, and OC in April were 4.1, 1.5, 0.5, and 1.3 µg m$^{-3}$, respectively, in the background model data. Although a careful evaluation is difficult because of the lack of aerosol chemical measurements, we speculated that the aerosols (other than OIN) were considerably low. Anthropogenic emissions might be biased for this city. The sources of emissions in residential/developing areas are principally anthropogenic; yet the residential emission factor for the emission inventory compilation is highly uncertain compared with the emission factors of power plants, industrial plants, and vehicles (Li et al., 2017). Chlorine and sodium are selected to represent sea-salt aerosols in WRF-Chem, yet the two concentrations were at very low concentrations in the model at Kashi. This was despite the fact that the Taklamakan Desert had many atmospheric halite particles, which were Cl- and Na-rich and accounted for 10% of the total particles in the desert (Okada and Kai, 2004).

For control variable design, our DA system modifies the chemical composition of each aerosol according to the BEC values. However, all the DA experiments showed that the PM$_{10}$ chemical fractions remain close to their background values (Figure 13). The low biases cannot be improved via DA because the aerosol chemical measurements were not available. In addition, the differences between DA_Esca and DA_Esca_Eabs were quite small (Figure 13c, e), indicating that assimilating the aerosol absorption coefficient did not enhance particle absorption (19.1 $^{-1}$ vs. 20.0 Mm$^{-1}$ in Table 3) in our system. The assimilation of the aerosol absorption coefficient alone (DA_Eabs) increased the percentage of BC to 7.1%, which was slightly higher than the 6.5% background BC. The first reason for the small changes in the aerosol chemical proportions is that the scattering aerosols (i.e., sulfate, nitrate, and ammonium), use the same refractive index and hygroscopicity parameter in WRF-Chem. Therefore, the AOD had virtually the same sensitivity to composition of each aerosol, and
assigned comparable modification to each composition. When assimilating the total quantities of aerosols (e.g., PM$_{2.5}$, PM$_{10}$, and AOD), it is difficult to distinguish different aerosol chemical contributions. Secondly, DA_Eabs increased the aerosol number concentration; meanwhile, the rising number concentration increased the scattering aerosols, which prevented a substantial rise in the BC fraction. Third, the concentrations of anthropogenic aerosols were lower than anticipated. They had comparable low background errors and could not be distinguished because of their small differences in BEC values. Overall, it seems that differences in aerosol chemical composition from assimilating the aerosol optical data are smaller than the difference in model setting (e.g., using other aerosol chemistry mechanisms, or using finer aerosol size bins). The assimilation of the total aerosol quantities cannot eliminate the intrinsic bias in aerosol composition. Thus, accurate aerosol chemistry and optical modules are crucial to attain a better background aerosol chemical data for DA analysis (Saide et al., 2020).

**Figure 13**

### 4.2 DA Impact on Aerosol Direct Radiative Forcing

Table 4 shows the instantaneous clear-sky ADRF in the background data and the analyses of DA_PM, and DA_PM$_{i}$, AOD. After the analyses, the DA effect (various DA frequencies for assimilating AOD and the surface particle concentrations) gradually faded away after restarting the model run. We therefore focused on the instantaneous radiative forcing values one hour after assimilating AOD data. This ensured that the comparison was based on similar analysis times and showed effective DA effects. As the dust was the predominant component, the ADRF in this section was closely equivalent to the dust radiative forcing.

Dust redistributes the energy between the land and the atmosphere. The atmosphere gains more shortwave energy as the dust particle absorption; the warming atmosphere also emits more longwave energy as it absorbs shortwave energy. The change in energy budget at the surface is correspondingly the opposite of that in the atmosphere. As shown in Table 4, the enhancements in surface cooling forces were slightly stronger than the atmospheric warming forcings. The differences between the surface forcing and atmospheric forcing indicate the ADRF at the top of the atmosphere (TOA). The TOA ADRF when assimilating the surface particle concentrations was enhanced by 12% in the shortwave, 83% in the longwave, and 6% in the net forcing values, and enhanced by 40%, 55%, and 38%, respectively, when assimilating the AOD. Apparently, assimilating PM$_{i}$ alone is not sufficient to accurately estimate the ADRF value. At Kashi, the total net clear-sky ADRF with assimilating surface particles and AOD were $-10.5$ Wm$^{-2}$ at the TOA, $+19.5$ Wm$^{-2}$ within the atmosphere, and $-30.0$ Wm$^{-2}$ at the surface, respectively, enhanced by 46%, 153%, and 100% respectively, compared to the background ADRF values. Because the AOD observation is only sporadically available because of cloud screening in retrieval data, the DA experiments still cannot eliminate the low bias in AOD in WRF-Chem. The ADRF values in the DA experiments are still likely to be lower than the plausible aerosol radiative forcing at Kashi.

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

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

The most abundant aerosol at Kashi in April 2019 was dust. The WRF-Chem model captured the main dust episodes, but only simulated half the monthly mean concentrations of PM$_{2.5}$ and PM$_{10}$. Furthermore, the model failed to capture the peak concentrations from a dust storm on 24 April. The aerosol scattering/absorption coefficients and AOD in the background data showed strong low biases and weak correlations with the observed levels. The DA systems did, however, effectively assimilate the surface particle concentrations, aerosol scattering coefficients, and AOD. Some deficiencies in the DA analysis were related to the forward model bias in transferring the aerosol mass concentrations to the aerosol optical parameter. Simultaneous assimilation of the PM$_{2.5}$ and PM$_{10}$ concentrations improved the model aerosol concentrations, with significant increases in the coarse particles; meanwhile, the analyzed AOD was 56% lower than observed levels. The assimilation of AOD significantly improved the AOD but overestimated the surface PM$_{10}$ concentration by a factor of at least two.

Assimilating the aerosol scattering coefficient improved the scattering coefficient in the analysis but overestimated the surface PM$_{10}$ concentration by 84%. It therefore seems that WRF-Chem underestimated the particle extinction efficiency. As a compensation, the DA system overestimated the aerosol concentration to fit the observed optical values, yielding overly high particle concentrations.

A notable problem was the assimilation of the absorption coefficient, which greatly overestimated the monthly mean values by a factor of at least four in the model parameters and yielded overly strong daily variations for the parameters. The aerosol absorption coefficient was improved but was still 21% lower than observed values. This defect was also apparent in the low particle absorption efficiency of WRF-Chem. The biases of the model in aerosol particle mixture and aged dust as well as the “missing” absorption of brown carbon,
accounted for the bias in absorption efficiency, which would have worsened the DA analysis when assimilating the absorption coefficient.

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

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

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

The limitations that necessitate further research include:

1. The binning strategy. The desired strategy should link the circulation flow and particle emission sources. A better hybrid DA coupled with the ensemble Kalman filter will be more effective for estimating the aerosol background error.

2. The observational error. This could be elaborated further. The \(\text{PM}_{10}\) included the anthropogenic coarse particles, which should be separated from the dust originating from the desert (Jin et al., 2019). We set the observation errors for \(\text{PM}_{10}\) and AOD to the conventional values. The observational errors of the nephelometer and aethalometer were slightly arbitrary in this study, necessitating further consideration.

3. The adjoint operator. This needs to be modified to assimilate the aerosol absorption coefficient or absorption AOD.

4. The DA system. Our revised DA system was based on four-size bin MOSAIC aerosols, but it can be extended to work with eight-size bin MOSAIC aerosols in WRF-Chem.

When assimilating aerosol optical data, the DA quality is strongly dependent on the forward model. The responses of our DA analysis to the bias and uncertainty in the forward aerosol 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.

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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 (µg 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. 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.
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$_{2.5}$ ($\mu$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 when assimilating the observed PM$_{2.5}$ and PM$_{10}$ (DA_PMx) and aerosol scattering coefficients (DA_Esca) at Kashi in April 2019.
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$_{2.5}$ (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_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) PM$_x$, (b) aerosol scattering coefficients ($E_{	ext{scat}}$), (c) aerosol absorption coefficient ($E_{	ext{abs}}$), 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 | 440, 675, 870, 1020 | $e = \sqrt{e_1^2 + e_2^2}$  
$e_1 = 1.5 + 0.75 \cdot PM_x$  
$e_2 = 0.5 \cdot e_1 \cdot \sqrt{\frac{d}{3000}}$  
$d$: 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)             | 261 (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  |
| Background | -7.8           | +11.3          | -19.1            | +0.6   | -3.6   | +4.2  |
| DA_PMx    | -8.7           | +21.7          | -30.4            | +1.1   | -7.4   | +8.5  |
| DA_PMx_AOD | -12.2          | +32.1          | -44.3            | +1.7   | -12.6  | +14.3 |

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