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Evaluations of Surface PM₁₀ Concentration and Chemical Compositions in MERRA-2 Aerosol Reanalysis over Central and Eastern China

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Abstract: The chemical composition dataset of Aerosol Reanalysis of NASA's Modern-Era Retrospective Analysis for Research and Application, version 2 (MERRAero) has not been thoroughly evaluated with observation data in mainland China due to the lack of long-term chemical components data. Using the 5-year data of PM₁₀ mass concentrations and chemical compositions obtained from the routine sampling measurements at the World Meteorological Organization the Global Atmosphere Watch Programme regional background stations, Jing Sha (JS) and Lin'an (LA), in central and eastern China, we comprehensively evaluate the surface PM₁₀ concentrations and chemical compositions such as sulfate (SO₄^{2−}), organic carbon (OC) and black carbon (BC) derived from MERRAero. Overall, the concentrations of PM₁₀, SO₄^{2−}, OC and BC from the MERRAero agreed well with the measurements, despite a slight and consistent overestimation of BC concentrations and a moderate and persistent underestimation of PM₁₀ concentrations throughout the study period. The MERRAero reanalysis of aerosol compositions performs better during the summertime than wintertime. By considering the nitrate particles in PM₁₀ reconstruction, MERRAero performance can be significantly improved. The unreasonable seasonal variations of PM₁₀ chemical compositions at station LA by MERRAero could be causative factors for the larger MERRAero discrepancies during 2016–2017 than the period of 2011–2013.

Keywords: MERRAero; PM₁₀; chemical compositions; evaluations; central and eastern China

1. Introduction

The Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2), is NASA's latest reanalysis of the atmospheric environment, with various environmental variables consistently in both temporal and spatial distributions [1]. The MERRA-2 combined the Goddard Earth Observing System, version 5, Earth system model (GEOS-5) [2,3], three-dimensional variational data assimilation, and Grid point Statistical Interpolation analysis system [4,5]. In this, "MERRAero" specifically refers to a reanalysis of aerosols in MERRA-2, including bias-corrected aerosol optical depth observations from the Moderate Resolution Imaging Spectroradiometers onboard the Terra and Aqua satellites [6], as well as the Goddard Chemistry-Aerosol-Radiation and Transport model [7,8]. The MERRAero dataset includes the concentrations of the five aerosol species of dust (DS),

sea salt (SS), sulfate (SO_4^{2-}), black carbon (BC), and organic carbon (OC) over the world with a resolution of 0.5° latitude by 0.625° longitude and 72 vertical layers extending up to 0.01 hPa (~ 80 km) [9]. In the GEOS-5 model, DS and SS emissions are classified into different diameter bins based on the relationship between surface properties and near-surface wind speed [10]. The emission inventories of other aerosol species and their precursors are considered during the simulation, in which, sulfate and carbonaceous aerosol emissions are derived from both natural and anthropogenic sources [1,11]. NASA's Quick Fire Emission Dataset provides the biomass burning emissions after 2009, including primarily OC and BC. Sulfur dioxide (SO_2 , the precursor of SO_4^{2-}) anthropogenic emissions come from the Emission Database for Global Atmospheric Research version 4.2 inventory in 2008 [12,13].

The differentiation of the aerosols' chemical compositions in MERRAero provides a valuable dataset for better studying various air quality issues around the world, especially for the regions lacking local particular matter (PM) observations due to unreliable or scarce monitoring [14]. Thus, many studies have emerged trying to estimate surface PM from MERRAero datasets in different regions of the world [12,14–17]. Some evaluation studies on surface PM of MERRAero were performed in recent years. In the United States, MERRAero $\text{PM}_{2.5}$ was closer to observation values during the summer while larger discrepancies were observed during the winter, because of the lack of nitrate emissions in MERRAero and an underestimation of carbonaceous emissions in the Western US [16]. In Europe, the evaluation of PM_{10} , $\text{PM}_{2.5}$, SO_4^{2-} and BC concentrations were generally reasonable except OC and SS, in which the wintertime bias is mainly contributed by the anthropogenic sources of PM unresolved by the simulation [15]. The MERRAero $\text{PM}_{2.5}$ in Taiwan was significantly underestimated, by 42% on average of 2005–2014, because emissions of anthropogenic PM and their precursors were largely uncertain in China [14].

The validation focusing on MERRAero-derived $\text{PM}_{2.5}$'s reliance and uncertainty by independent measurements has been conducted spatially and temporally in China to support its applications related to climate-aerosol interactions [18–21]. Generally, MERRAero well captures the spatial distribution and seasonal variations of $\text{PM}_{2.5}$ mass concentration in China mainland. However, MERRAero produced lower daily mean $\text{PM}_{2.5}$ concentrations with outstanding bias for high ground $\text{PM}_{2.5}$ ($>75 \mu\text{g m}^{-3}$) [19]. Significant underestimation of the $\text{PM}_{2.5}$ mass concentration in MERRAero was revealed across China mainland according to a 5-year evaluation, especially in Central China by $34.6 \mu\text{g m}^{-3}$, followed by $19.8 \mu\text{g m}^{-3}$ in East China and $9.1 \mu\text{g m}^{-3}$ in South China [20]. Additionally, the bias also exhibited seasonal dependence. The largest biases were observed in winter. Due to the low intensity and weak variations of emission inventory as well as the absence of nitrate, the magnitude and variability of $\text{PM}_{2.5}$ in MERRAero are both underestimated [20]. Compared with the widely assessments of $\text{PM}_{2.5}$ product in MERRAero, the chemical speciation of MERRAero has not been thoroughly evaluated with observation data in mainland China due to the lack of long-term chemical components data. In this study, five years of aerosol sample data collected at the World Meteorological Organization's (WMO) Global Atmosphere Watch (GAW) programme's regional background stations, Jing Sha (JS) and Lin'An (LA), in central and eastern China, will be used to evaluate the aerosol concentrations and chemical speciation of MERRAero. Three observation-based methods for reconstructing nitrate will be proposed and evaluated to offset the key absence of MERRAero nitrate. The evaluation results for surface aerosol concentration and especially chemical compositions are necessary for improving MERRAero's applicability in highly polluted regions in China, especially for improving its applicability to the regional background levels of aerosol compositions.

The paper is organized as follows: the observation measurements of chemical components and the methods used for reconstruction and validation are introduced in Section 2; the seasonal and monthly variability of PM_{10} and chemical compositions estimated by the MERRAero at two regional background stations are comprehensively validated in Section 3.1; three observation-based methods for making up the nitrate pollutants in the MERRAero

are performed and evaluated in Section 3.2; the uncertainty analysis and discussion are presented in Section 3.3; and the assessments are summarized in Section 4.

2. Materials and Methods

2.1. Observation Sites and PM_{10} Sample Analysis

Aerosol samples were collected from 2011 to 2013 and 2016 to 2017 at the WMO GAW atmospheric regional background stations, JS station in Hubei province and LA station in Zhejiang province, representing the regions of Twin Lake Basin and Yangtze River Delta over central and eastern China with high aerosol loading (Figure 1). JS station ($29^{\circ}38'N$, $114^{\circ}12'E$, 750 m a.s.l.) is located in the east wing of the Twin Lake Basin region, about 105 km south of Wuhan, the capital city of Hubei province. Situated on the peak of Monk mountain, JS station is about 30 km east of Chongming county, which is considered to represent the regional background atmospheric components over the Twin Lake Basin region. LA station ($30^{\circ}18'N$, $119^{\circ}44'E$, 138.6 m a.s.l.) is located in the rural area of Zhejiang Province, which is close to rapidly developing regions in Jiangsu province and the mega-city Shanghai. The regional background characteristics of the atmospheric components in Yangtze River Delta can be well represented by LA station [22].

Figure 1. Spatial distributions of aerosol optical depth from the Moderate Resolution Imaging Spectroradiometers averaged over 2011–2020 and the locations of two background observation stations (red dots) JingSha (JS) and Lin'An (LA) in central and eastern China.

There were 499 total aerosol samples for LA station and 317 aerosol samples for JS station collected in the five years over 2011–2013 and 2016–2017. The detail information for aerosol samples analysis is summarized in Table 1. Aerosol samples were collected every two or three days by using a MiniVolTM air sampler (Airmetrics, Springfield, OR, USA), operating at a sampling flow rate of 5 L min^{-1} for 24 h from 09:00 a.m. to 09:00 a.m. (Beijing time) the next day. The 47 mm Whatman quartz microfibre filters (QMA) pre-fired at 850°C for 3 h were used for the sampling.

The aerosol mass concentrations of PM_{10} were obtained by gravimetric analysis in the Laboratory of the Chinese Academy of Meteorological Sciences with a microbalance (Sartorius, Germany). The water-soluble ions, including nine inorganic ions (F^{-} , Cl^{-} , NO_3^{-} , SO_4^{2-} , NH_4^{+} , K^{+} , Na^{+} , Ca^{2+} , Mg^{2+}), were analyzed by using ion chromatography (Dionex 3000 IC and atomic absorption spectrophotometry HITACHI 180-70) at the Laboratory of the Chinese Academy of Meteorological Sciences. The elemental carbon (EC) and organic

carbon (OC) were analyzed by using a DRI model 2001A EC/OC analyzer. The methods used were the same as described in Zhang et al. [23].

Table 1. The measurement methods and instruments used in the aerosol sample analysis.

Aerosol Samples	Measurement	Instrument
PM ₁₀	Gravimetric analysis	MiniVol™ air sampler and the 47 mm Whatman quartz microfibre filters
Inorganic ions (F [−] , Cl [−] , NO ₃ [−] , SO ₄ ^{2−} , NH ₄ ⁺ , K ⁺ , Na ⁺ , Ca ²⁺ , Mg ²⁺)	Ion chromatography	Dionex 3000 IC and atomic absorption spectrophotometry
The elemental carbon (EC) and organic carbon (OC)	Thermal/Optical method	DRI 2001A EC/OC analyzer

2.2. PM₁₀ Reconstruction and Evaluation Methods

The MERRAero provides the concentration of five PM species on the 3-hourly basis: SO₄^{2−}, OC, BC, DS and SS. Based on the available aerosol chemical speciation measurements, PM mass can be reconstructed with equations [24]. The total concentration of PM₁₀ (PM with diameter < or = 10 µm) is estimated as follows:

$$[\text{PM}_{10}] = 1.375 \times [\text{SO}_4^{2-}] + 1.8 \times [\text{OC}] + [\text{BC}] + [\text{DS}_{10}] + [\text{SS}_{10}] \quad (1)$$

We use brackets to denote concentrations. Assuming SO₄^{2−} is fully neutralized by ammonium (NH₄⁺) in the form of ammonium sulfate ((NH₄)₂SO₄), [SO₄^{2−}] is multiplied by 1.375 to account for sulfate. Other organic compounds found in particulate organic matter (POM) can be represented with [OC] multiplied by 1.8 [14–16]. DS₁₀ and SS₁₀ can be derived from the first three size bins in MERRAero's DS and SS.

The lack of the concentration of nitrate particles in MERRAero can be problematic in PM mass construction, as nitrate is growing into a bigger proportion in aerosols across China [16,25]. Thus, we use the modified Equation (1) to reconstruct PM₁₀ mass concentrations with nitrate included as follows:

$$[\text{PM}_{10}] = 1.375 \times [\text{SO}_4^{2-}] + 1.29 \times [\text{NO}_3^-] + 1.8 \times [\text{OC}] + [\text{BC}] + [\text{DS}_{10}] + [\text{SS}_{10}] \quad (2)$$

Assuming NO₃[−] is fully neutralized by NH₄⁺ in the form of ammonium nitrate (NH₄NO₃), we can estimate nitrate with 1.29 × [NO₃[−]]. In this study, the concentration of [NO₃[−]] was estimated based on the ratio of observed [NO₃[−]] to [SO₄^{2−}]. There is mineral composition data available during 2016 to 2017 in two sites for obtaining a reasonable estimation of [DS₁₀] and [SS₁₀] [26], and the DS and SS species in MERRAero are also evaluated.

With sulfate data available in MERRAero, the nitrate concentration can be derived through the ratio of observed [NO₃[−]] to [SO₄^{2−}]. The abovementioned Equation (2) is used to reconstruct PM₁₀ with considering nitrate. Here we use three methods to estimate the MERRAero-nitrate based on three timescale ratios of observed [NO₃[−]] to [SO₄^{2−}] as Equation (3): (1) “PM₁₀ (yearly ratio)” based on observed [NO₃[−]]/[SO₄^{2−}] on a yearly basis during studying period; (2) “PM₁₀ (monthly ratio)” based on observed [NO₃[−]]/[SO₄^{2−}] on a monthly basis; (3) “PM₁₀ (daily ratio)” based on observed [NO₃[−]]/[SO₄^{2−}] on a daily basis. The monthly ratio of observed [NO₃[−]] to [SO₄^{2−}] increased during the past few years (Figure 2), indicating the inevitable importance to induce MERRAero-nitrate due to its growing proportion in aerosols.

$$\text{MERRAero } [\text{NO}_3^-] = \text{Observed } [\text{NO}_3^-] / [\text{SO}_4^{2-}] \times \text{MERRAero } [\text{SO}_4^{2-}] \quad (3)$$

Figure 2. The monthly averaged ratio of observed $[\text{NO}_3^-]$ to $[\text{SO}_4^{2-}]$ during two periods over 2011–2013 and 2016–2017 in (a) LA and (b) JS.

To quantify MERRAero's accuracy, the performance statistics we used include the mean fraction $\bar{F} = \bar{C}_s / \bar{C}_o$, (C_s : simulated concentration; C_o : observed concentration), the mean bias $\bar{B} = \bar{C}_s - \bar{C}_o$, the standard deviation of the bias (SD-B) and the correlation coefficient (R). Despite being widely applied, the correlation coefficient R is a criticized index for evaluating model performance since it does not directly compare the simulated results with observed data [27]. Therefore, we adopt a rigorous index to evaluate the MERRAero performance. The proportion of simulated data which falls within a factor of 2 of the observed data (FAC2, i.e., the proportion of the data which satisfies $0.5 \leq \bar{C}_s / \bar{C}_o \leq 2.0$), such a method can avoid the influence of extreme values and errors. It is considered the simulation performance to be reasonably good if $\text{FAC2} > 0.50$ [28]. Besides, the index of agreement (IOA) was also used to assess the MERRAero performance in PM_{10} against the measurements.

$$\text{IOA} = 1 - \frac{\sum_{i=1}^N (C_{s,i} - C_{o,i})^2}{\sum_{i=1}^N (|C_{s,i} - \bar{C}_s| + |C_{o,i} - \bar{C}_o|)^2} \quad (4)$$

where N is the total number of the samples used for comparisons. The IOA is a standard measure of the degree of model accuracy and ranges from 0 to 1, with 1 showing perfect agreement of the prediction with the observation [29–31].

3. Results

3.1. Evaluation of Aerosol Species

The performance statistics of MERRAero aerosol compositions at two regional background stations, LA and JS, are summarized in Tables 2 and 3. For the period 2011 to 2013, the observed average PM_{10} concentration at LA and JS reached $94.81 \mu\text{g m}^{-3}$ and $85.33 \mu\text{g m}^{-3}$ respectively, which is significantly high compared to $16.50 \mu\text{g m}^{-3}$ in Europe (averaged with 55 sites) [15]. On average, MERRAero underestimates $[\text{PM}_{10}]$ and $[\text{OC}]$ at LA with the mean bias -33.97 and $-1.38 \mu\text{g m}^{-3}$ by a factor of 0.64 and 0.88, respectively. $[\text{BC}]$, on the other hand, is overestimated by a factor of 1.36 with a discrepancy of $1.12 \mu\text{g m}^{-3}$. $[\text{SO}_4^{2-}]$ is estimated very well with the mean fraction $\bar{F}$ around 1.00 and has only $0.05 \mu\text{g m}^{-3}$ average bias compared to Europe [15]. Compared to LA, JS underestimates $[\text{PM}_{10}]$ and $[\text{SO}_4^{2-}]$ respectively with the factors of 0.73 and 0.87 in the discrepancy of $-23.29 \mu\text{g m}^{-3}$ and $-2.81 \mu\text{g m}^{-3}$, while $[\text{OC}]$ and $[\text{BC}]$ are overestimated compared to observations by $1.06 \mu\text{g m}^{-3}$ and $1.42 \mu\text{g m}^{-3}$. Overall, the PM_{10} concentrations and chemical compositions at LA and JS site are generally better reproduced by the MERRAero than that at Europe [15] based on statistical parameters. For the period 2016–2017, the observed average PM_{10} concentration at LA and JS is reduced to $61.37 \mu\text{g m}^{-3}$ and $58.44 \mu\text{g m}^{-3}$ respectively due to the strict and comprehensive air pollution control policies implemented by the government from 2013. MERRAero largely underestimates $[\text{PM}_{10}]$ and all the PM components in LA. The possible reasons for the obvious discrepancy of MERRAero and observation in LA during 2016–2017 will be discussed in the

Section 3.3. The $[\text{SO}_4^{2-}]$ and $[\text{PM}_{10}]$ in JS are relatively estimated well with discrepancy of $3.99 \mu\text{g m}^{-3}$ and $-4.00 \mu\text{g m}^{-3}$. $[\text{DS}_{10}]$ in both LA and JS is underestimated. In general, MERRAero performance in the second period 2016–2017 is less favorable than the first period 2011–2013.

Table 2. Statistics of the Aerosol Reanalysis of NASA’s Modern-Era Retrospective Analysis for Research and Application, version 2 (MERRAero) performance against the observations at LA and JS over 2011–2013.

	LA				JS			
	OC	BC	SO_4^{2-}	PM_{10}	OC	BC	SO_4^{2-}	PM_{10}
n	302	302	291	295	156	156	156	156
AOC * ($\mu\text{g m}^{-3}$)	11.38	3.16	16.78	94.81	8.81	2.24	20.89	85.33
$\bar{F}$	0.88	1.36	1.00	0.64	1.12	1.63	0.87	0.73
$\bar{B}$ ($\mu\text{g m}^{-3}$)	−1.38	1.12	0.05	−33.97	1.06	1.42	−2.81	−23.29
SD-B ($\mu\text{g m}^{-3}$)	6.06	2.22	9.52	54.71	5.86	1.98	10.33	45.44
R	0.41	0.48	0.53	0.45	0.45	0.56	0.62	0.55
IOA	0.62	0.64	0.72	0.58	0.63	0.62	0.76	0.63
FAC2	0.81	0.69	0.82	0.66	0.79	0.62	0.81	0.81

* AOC stands for “average observed concentration”.

Table 3. Statistics of the Aerosol Reanalysis of NASA’s Modern-Era Retrospective Analysis for Research and Application, version 2 (MERRAero) performance against the observations at LA and JS over 2016–2017.

	LA						JS					
	OC	BC	SO_4^{2-}	PM_{10}	DS_{10}	SS_{10}	OC	BC	SO_4^{2-}	PM_{10}	DS_{10}	SS_{10}
n	197	186	197	197	197	193	161	161	161	161	161	157
AOC * ($\mu\text{g m}^{-3}$)	6.64	3.09	9.57	61.37	20.03	0.50	4.71	2.30	10.27	58.44	18.28	0.55
$\bar{F}$	0.61	0.64	0.81	0.52	0.58	0.54	1.79	1.49	1.39	0.93	0.83	1.90
$\bar{B}$ ($\mu\text{g m}^{-3}$)	−2.61	−1.11	−1.77	−29.53	−8.45	−0.23	3.72	1.12	3.99	−4.00	−3.09	0.49
SD-B ($\mu\text{g m}^{-3}$)	6.02	2.65	13.33	54.50	18.96	0.50	4.76	2.37	7.58	31.12	23.17	1.04
R	−0.12	0.02	−0.15	−0.03	0.22	−0.05	0.55	0.36	0.46	0.45	0.17	−0.08
IOA	0.34	0.41	0.24	0.40	0.49	0.39	0.58	0.53	0.61	0.66	0.37	0.17
FAC2	0.43	0.42	0.24	0.37	0.40	0.28	0.58	0.64	0.74	0.83	0.45	0.34

* AOC stands for “average observed concentration”.

All species at LA and JS have reasonably low $\bar{B}$ values (Tables 2 and 3). However, there is quite a lot of scattering in the data, as reflected by the values SD-B, which are much larger than their respective average bias, accompanied with low R values. The data scattering is noticeable in Figure 3, especially for the PM species at the LA site. Even though, the bulk of data are still well simulated by MERRAero with good linear fitting results. Since all the FAC2 factors at LA and JS exceed 60% (Table 2), it is considered that the MERRAero performance is reasonably good. With the FAC2 values > 80%, $[\text{OC}]$ and $[\text{SO}_4^{2-}]$ in LA site, and $[\text{PM}_{10}]$ and $[\text{SO}_4^{2-}]$ in JS site, are all overall very well estimated. Except for PM_{10} in LA, other species have relatively higher FAC2 than Europe’s values [15], and reasonable IOA indicating good data consistency with observations. It is worth noting that PM_{10} evaluation results in Tables 2 and 3 were calculated with Equation (1) without consideration of nitrate particles.

Figure 3. Scatter plots between observations and reanalysis of PM₁₀ species ($\mu\text{g m}^{-3}$) at LA and JS for the first period 2011–2013 with liner fitting lines (solid lines) passing 99% significance level and 1:1 line (dash lines) between observations and the MERRAero.

Looking into the monthly averaged PM concentration in five years' span (Figure 4), there are considerable discrepancies of [OC] and [SO₄²⁻] at LA site in 2011 and 2016.

The underestimation of PM_{10} is likely caused by a combination of $[SO_4^{2-}]$ and $[OC]$ underestimations. For JS site, the lesser extent of monthly variation in $[SO_4^{2-}]$ seems to be the major contributor for PM_{10} inconsistency. The most obvious difference between LA and JS existed in the MERRAero performance of $[SO_4^{2-}]$ (Tables 2 and 3 and Figure 4). MERRAero overestimated $[SO_4^{2-}]$ with the lesser amplitude of monthly variation comparing with the observation in JS. The site JS is located on the remote mountaintop at an altitude of 750 m a.s.l. with the very low anthropogenic emissions of air pollutants and is often influenced by relatively clean air from above the boundary layer when the height of boundary layer is lower in cold season [32]. Air pollutants over the remote mountain are mostly transported from the high emission regions under a favorable synoptic system [33], which could lead to the significant monthly variations of air pollutants. Furthermore, the spatial resolution of 0.5° latitude by 0.625° longitude in the MERRAero data is too coarse to accurately present the mountainous topography, which could be responsible for the overestimation and gentle variations of $[SO_4^{2-}]$ in the site JS over the remote mountain in central China, while the relative reasonable $[SO_4^{2-}]$ was produced in LA over the plain of East China with the relatively high anthropogenic emissions of air pollutants. In both sites, MERRAero's $[SO_4^{2-}]$ in the second period, 2016 and 2017, shows larger discrepancy and overestimation. The use of a constant inventory of SO_2 emissions from 2008 [13] and anthropogenic carbonaceous in Aero Com Phase II dataset (HCA0v1) from 2006 [34] in MERRAero are problematic in the long-term simulation, despite that the use of 3DVar technique could partially improve the simulation accuracy [14]. The emissions are constantly changing with time, especially in China where the strict and comprehensive air pollution control policies were implemented by the government from 2013 [25]. Even though the bias of PM_{10} species existed along the study period with the regional differences, the decreasing trend of PM_{10} and chemical components since 2013 are reproduced by MERRAero, and there is still a potential to improve the performance of reconstructed PM_{10} by considering the nitrate contribution to the simulation results.

Figure 5 compares the monthly averaged PM_{10} concentration and chemical compositions reproduced by MERRAero with observations for the first period 2011–2013. Overall, there is less discrepancy between the observed and reanalyzed concentrations for all aerosol species in the summertime than in winter. The LA site presents more accurate seasonal variations and fewer concentration discrepancies with observations for all PM species than JS site. Except for the underestimation of MERRAero's $[OC]$ in winter, $[SO_4^{2-}]$ and $[BC]$ in MERRAero both have relatively good agreements with the observations on monthly basis. MERRAero's performance is less favorable in PM_{10} , showing persistent underestimation throughout most of the months. The PM_{10} reconstruction of MERRAero in Equation (1) is lack of the concentration of nitrate particles, which could be the reason for the consistent underestimation in reconstructed PM_{10} during the study period. In the following Section 3.2, we will testify and compensate the contribution of nitrate particles to reconstructed PM_{10} .

It is noted that a large discrepancy exists between MERRAero and observations in November (N) and December (D) at JS site. The reason is mainly because that the JS station is located on the mountaintop (750 a.s.l.) and often influenced by clean air from the lower free troposphere when the boundary layer is low with wind coming from north west [32]; however, such a case might not be well represented by the MERRAero, and result in the overestimation of chemical compositions from MERRAero in November and December at JS (see Figure 5).

Figure 4. Monthly variations of PM species (a,e) organic carbon (OC), (b,f) sulfate (SO_4), (c,g) black carbon (BC) and (d,h) PM_{10} from observations (black lines) and MERRAero (red lines) at sites LA (left column) and JS (right column) for the two periods over 2011–2013 and 2016–2017.

Figure 5. Seasonal variations of observations (black lines) and MERRAero reanalysis (red lines) for (a) OC, (b) SO_4^{2-} , (c) BC and (d) PM_{10} at LA (solid lines) and JS (dash lines) respectively.

3.2. Improvement of PM₁₀ Reconstruction with Including Nitrate

With a growing proportion of nitrate in aerosols over China, the lack of the concentrations of nitrate particles in MERRAero dataset can make up a considerable portion of the total [PM₁₀] and [PM_{2.5}] loss [16,25]. Performance statistics of reconstructed MERRAero's [PM₁₀] based on different timescale nitrate inclusion methods compared with no-nitrate [PM₁₀] for the first period 2011 and 2013 are summarized in Table 4. MERRAero underestimated [PM₁₀] by a factor of 0.64 with the mean bias of $-33.97 \mu\text{g m}^{-3}$ in LA and 0.73 with the mean bias of $-23.29 \mu\text{g m}^{-3}$ in JS without considering nitrate particles. By including nitrate in PM₁₀ (yearly ratio), the averaged underestimations were significantly improved with the mean bias moving from -33.97 to $-24.79 \mu\text{g m}^{-3}$ in LA and from -23.29 to $-14.42 \mu\text{g m}^{-3}$ in JS, and the mean fraction of simulation and observation $\bar{F}$ can be enhanced from 0.64 up to 0.74 at LA and raised from 0.73 to 0.83 at JS. The mean bias $\bar{B}$ of three methods (PM₁₀ (yearly ratio), PM₁₀ (monthly ratio), PM₁₀ (daily ratio)) were quite close, implying a limit could exist in [PM₁₀] improvement by adopting the fine temporal resolution of observed $[\text{NO}_3^-]/[\text{SO}_4^{2-}]$ (i.e., from yearly to monthly, and then daily) to estimate MERRAero nitrate concentration. Figure 6 shows that the scattering in the reconstructed [PM₁₀] became less obvious from PM₁₀ (yearly ratio) to PM₁₀ (daily ratio) in both LA and JS, which is also confirmed by the declines in SD-B, increases in R, IOA and FAC2 (Table 4). It is worth noting that it is acceptable to include the nitrate concentration with our method for PM₁₀ construction by averaged observed $[\text{NO}_3^-]/[\text{SO}_4^{2-}]$ in a period of time considering the difficulties of high temporal-spatial resolution in chemical speciation observation. The nitrate-induced efforts would significantly enhance the MERRAero's applicability in highly polluted regions, especially with the relatively high nitrate concentrations in ambient air.

Table 4. Statistics of MERRAero PM₁₀ reconstruction performance with different timescale nitrate estimation methods for the first period from 2011 to 2013 at LA and JS.

	LA				JS			
	PM ₁₀ (No Nitrate)	PM ₁₀ (Day)	PM ₁₀ (Month)	PM ₁₀ (Year)	PM ₁₀ (No Nitrate)	PM ₁₀ (Day)	PM ₁₀ (Month)	PM ₁₀ (Year)
n	295	285	295	295	156	156	156	156
AMC * ($\mu\text{g m}^{-3}$)	60.80	69.83	70.63	70.02	64.32	71.42	73.16	73.89
$\bar{F}$	0.64	0.735	0.744	0.738	0.73	0.834	0.826	0.831
$\bar{B}$ ($\mu\text{g m}^{-3}$)	-33.97	-25.26	-24.26	-24.79	-23.29	-14.27	-14.80	-14.42
SD-B ($\mu\text{g m}^{-3}$)	54.71	48.39	49.49	50.22	45.44	42.89	41.26	41.00
R	0.45	0.53	0.48	0.46	0.55	0.53	0.57	0.57
IOA	0.58	0.67	0.64	0.62	0.63	0.68	0.69	0.69
FAC2	0.66	0.79	0.76	0.75	0.81	0.87	0.86	0.88

* AMC stands for "average modeled concentration".

The daily averages of PM₁₀ observations, and MERRAero-reconstructed PM₁₀ with and without daily nitrate included during study periods (2011–2013 and 2016–2017) are demonstrated in Figure 7. High PM₁₀ concentration events are not well detected by MERRAero, especially during 2016 and 2017 in LA. For JS station, several months of observations are lacked during 2011 and 2013 due to technical issues. Although MERRAero underestimates the PM₁₀ compared with the observations, including nitrate in PM₁₀ could significantly improve the model performance in the secondary pollution circumstances.

Figure 6. Scatter plots between PM₁₀ observations and MERRAero PM₁₀ ($\mu\text{g m}^{-3}$) at sites (a) LA and (b) JS over 2011–2013. Reconstructed MERRAero PM₁₀ without nitrate particle included are represented as grey dots. Reconstructed MERRAero PM₁₀ based on yearly, monthly and daily ratio of observed $[\text{NO}_3^-]$ to $[\text{SO}_4^{2-}]$ are indicated as light pink, pink and red dots, respectively. Dash lines are $y = x$.

Figure 7. Daily averages of the MERRAero PM₁₀ without nitrate (gray contour), PM₁₀ observations (black dots) and the MERRAero PM₁₀ with daily reconstructed nitrate (red dots) at sites LA and JS during 2011–2013 and 2016–2017.

3.3. Bias Analysis and Discussion

The statistical analysis results in Table 3 revealed the obvious discrepancy of MERRAero products in LA during 2016–2017. We examined the individual seasonal variations of MERRAero as well as observed PM chemical components during the 5-year study period (Figure 8). For the years of 2016 and 2017, the MERRAero analysis showed unreasonable seasonal variation at LA with higher concentrations in summer and lower concentrations in winter, which is not in agreement with the observed high concentrations during winter. The possible reasons could be the uncertainty of emissions, the error of data assimilation, and the simulation bias of meteorology, which lead to the significant underestimation of wintertime PM chemical components. Meteorology exerts impact significantly on PM formation and accumulation. In terms of the simulation bias of meteorology, the overestimation of near-surface winds and boundary layer height could cause the PM underestimation with stronger diffusion in the atmosphere [5]. More precipitation enhances the PM removals. Lower humidity and air temperature suppress PM formation. The reasons for the discrepancy in MERRAero analysis in LA during 2016–2017 still need more research.

Figure 8. Seasonal variations of OC, SO_4^{2-} and BC in MERRAero reanalysis (left column) and observation (right column) in LA during the 5-year study period.

According to the averaged aerosol concentrations in observation and MERRAero during the first period 2011 to 2013, the contributions of different compositions apart from coarse particles to the PM_{10} mass concentrations at two sites are identified in Table 5. The major identified aerosol types in PM_{10} at LA and JS both were sulfate (as $(\text{NH}_4)_2\text{SO}_4$), particulate organic matter (POM), nitrate (as NH_4NO_3) and BC, with their contributions of 24%, 22%, 11% and 3% in LA, and 34%, 19%, 10% and 3% in JS, respectively. The MERRAero PM_{10} based on daily reconstructed nitrate were similar from observations, with sulfate (33%), POM (26%), nitrate (13%) and BC (6%) in LA and sulfate (38%), POM (26%),

nitrate (10%) and BC (5%) in JS. The reconstructed PM_{10} with nitrate included is still lower than the observation by around $25 \mu g m^{-3}$ in LA and $14 \mu g m^{-3}$ in JS.

Table 5. Averages of observed and MERRAero aerosol concentrations at LA and JS over 2011–2013.

	Observation (LA)		MERRAero (LA)		Observation (JS)		MERRAero (JS)	
	Con.	Pro	Con.	Pro	Con.	Pro	Con.	Pro
PM_{10}	94.81		69.83		85.33		71.42	
$(NH_4)_2SO_4$	23.06	24%	23.38	33%	28.71	34%	26.85	38%
NH_4NO_3	10.16	11%	9.03	13%	9.04	10%	7.10	10%
POM	20.49	22%	18.05	26%	15.86	19%	18.46	26%
BC	3.16	3%	4.33	6%	2.24	3%	3.83	5%
DS_{10}			12.01	17%			14.03	20%
SS_{10}			3.03	4%			1.14	2%

The observation analysis of aerosol types at LA indicated sulfate and soil aerosol contributes a significant proportion [20]. The location of two observation stations is in the inland with less contribution of coarse sea salt aerosol. In the Section 3.1, we attribute the underestimation of sulfate to the constant inventory of SO_2 emissions used. Here we focus on discrepancy in the soil aerosol. According to Figure 9a–d, observed DS_{10} rank as the primary components in PM_{10} during 2016–2017 both in LA (31%) and JS (31%). The contribution of aerosol types in PM_{10} is relatively well reproduced by MERRAero in LA. As for JS station, MERRAero underestimated the proportion of DS_{10} by 10%. The model generally had difficulty simulating dust concentrations due to both limitations of the windblown dust emission module and uncertainties in the anthropogenic fugitive dust emissions inventories [35]. The discrepancy of MERRAero's [PM_{10}] could result from the underestimation of coarse particles originating from natural sources and anthropogenic emissions. Natural dust emissions in MERRAero depend on the strong winds exceeding the threshold values, and the threshold wind-speed for emission also depends on soil moisture and other surface properties over the desert regions [36]. The location of observation stations is relatively far away from the large desert area in northwestern China. Thus, in central and eastern China with intensive human activities, the emissions of anthropogenic fugitive dust could be an important factor for the PM_{10} concentrations in the sites LA and JS. The discrepancies of DS may lay on excluding the anthropogenic fugitive dust emissions, which is a challenge for further modelling investigation.

Figure 9. Contributions of the different types of aerosols to PM_{10} in observation (left column) and MERRAero (right column) at sites LA and JS over 2016–2017, respectively.

4. Conclusions

We evaluated MERRAero reanalysis of PM₁₀ concentrations and chemical compositions with five-year observation data at two WMO GAW regional background stations, Jingsha and Lin'An, in central and eastern China. The chemical compositions of MERRAero have been thoroughly validated with observation data in mainland China. Three methods introducing nitrate to MERRAero PM₁₀ in yearly, monthly and daily timescales were compared to assess the PM₁₀ improvements. Such efforts could enhance the MERRAero's applicability related to climate-aerosol interactions in pollution regions, especially with the relatively high nitrate concentrations in ambient air. The main conclusions are summarized as follows:

- (a) Overall, the MERRAero provides the overall good analysis products of [PM₁₀], [SO₄^{2−}], [OC] and [BC] in two regional sites in central and eastern China. The average bias, IOA and, most importantly, the rigorous index FAC2 presented favorable results.
- (b) The evaluation of MERRAero [SO₄^{2−}] is relatively more encouraging in LA site than JS site, with a small average bias of 0.05 μg m^{−3}. MERRAero consistently overestimated [BC] by a factor of 1.35 on average but contributed relatively little to total PM₁₀ concentration. Underestimations of MERRAero [PM₁₀] and [OC] are most substantial in wintertime than summertime.
- (c) Compared with the first period 2011–2013, MERRAero performance on PM₁₀ and components is less favorable with larger discrepancy during the second period 2016–2017, even though the decreasing trend of PM₁₀ since 2013 is reproduced by MERRAero. The possible reasons may come from the uncertainty in sulfate and soil aerosol emissions.
- (d) By introducing nitrate based on the ratio of observed [NO₃[−]] to [SO₄^{2−}] from three different timescales (yearly, monthly and daily), the PM₁₀ from the MERRAero were improved substantially. The yearly ratio of [NO₃[−]] to [SO₄^{2−}] observation has the similar improvement in the MERRAero's applicability in central and eastern China with the daily ratio of [NO₃[−]] to [SO₄^{2−}]. It is essential to include the nitrate for MERRAero PM₁₀ and PM_{2.5} reconstruction for further long-term aerosol analysis in China region.
- (e) The MERRAero performances of PM₁₀ components of [SO₄^{2−}], [OC] and [BC] varied temporally in central and eastern China, thus emission inventories of MERRAero need to be updated timely in order to keep up with the recent emission mitigation nowadays, especially in China region.

Our results revealed an encouraging prospect of using MERRAero dataset in broad and comprehensive aerosol analysis. Still, more assessments about the MERRAero chemical components are needed by more recent PM observation data in different regions of China. For example, coarse particulates evaluation in northwestern China and fine chemical components in central China are necessary to facilitate the MERRAero dataset's improvements and application.

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