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Original Article

Evaluation of PM surface concentrations simulated by Version 1 of NASA's MERRA Aerosol Reanalysis over Europe



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ABSTRACT

This article evaluates the concentrations of particulate matter (PM) and some of its chemical speciation such as sulfate, organic carbon, black carbon and sea salt particles simulated at the surface by Version 1 of the Aerosol Reanalysis of NASA's Modern-Era Retrospective Analysis for Research and Application (MERRAero) over Europe. Measurement data from the European Monitoring and Evaluation Programme database were used. The concentrations of coarse PM (PM₁₀), fine PM (PM_{2.5}), sulfate and black carbon particles are overall well simulated, despite a slight and consistent overestimation of PM₁₀ concentration, and a slight and consistent underestimation of PM_{2.5} and sulfate concentrations throughout most of the year. The concentration of organic carbon was largely underestimated, especially in winter, caused by two specific monitoring stations in Italy, resulting in an overall poor performance for this particular species. After removing these two stations from the sample, the evaluation of OC substantially improved but an underestimation in winter remained. Carbon emissions originating from anthropogenic sources, such as residential wood burning in winter, unresolved by MERRAero provide a plausible explanation for this discrepancy. The evaluation of PM_{2.5}, sulfate and organic carbon concentrations improved during the summer. The concentration of fine sea salt particles was consistently and largely overestimated, but contributes relatively little to total PM_{2.5} concentration.

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1. Introduction

Many years ago, NASA introduced the first version of the Modern-Era Retrospective Analysis for Research and Application (MERRA), a reanalysis tool designed to simulate the hydrological cycle since the satellite era for climate research purposes (Rienecker et al., 2011). MERRA integrates observations from NASA's Earth Observing System satellites and the 5th version of the Goddard Earth Observing System (GEOS-5) atmospheric model and data assimilation system (Rienecker et al., 2008).

Recently, MERRA was expanded to assimilate bias-corrected aerosol optical depth (AOD) observations from the Moderate Resolution Imaging Spectroradiometers (MODIS, Remer et al.,

2005) on board the Terra and Aqua satellites, and the Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) model (Chin et al., 2002). GOCART simulates the sources, sinks, transport and concentration of sulfate (SO₄), organic carbon (OC), black carbon (BC), dust (DS) and sea salt (SS) aerosols from the meteorological fields provided by GEOS-5 and emission inventories of different air pollutants. DS and SS emissions are a function of surface properties and wind speed at the surface, and their respective concentrations are classified in different diameter bins. Sources of other species are simulated from emission inventories, including their precursors. Sulfur dioxide (SO₂, the precursor of SO₄) anthropogenic emissions are input from the Emission Database for Global Atmospheric Research (EDGAR) version 4.1 inventory from 2005 and biomass burning emissions (primarily OC and BC) are input from the NASA Quick Fire Emission Dataset (QFED) version 2.1 (Buchard et al., 2015). Version 1 of MERRA's Aerosol Reanalysis is labeled "MERRAero."

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MERRAero simulates the concentrations of the five aerosol species listed in the previous paragraph all over the world with a resolution of 0.5° latitude by 0.625° longitude and 72 vertical layers (from the surface to 80 km) between 2002 and 2015 (Buchard et al., 2015). Considering that airborne particulate matter (PM) impacts public health and visibility differently depending on its size and its chemistry (e.g., Laden et al., 2000; Schwartz and Neas, 2000; Groblicki et al., 1981), MERRAero's ability to differentiate chemical speciation of aerosols is a significant improvement for studying a broad range of air quality issues around the world since very few monitoring networks make such an identification of local PM and its speciation observations, but particularly in regions with unreliable or scarce monitoring.

MERRAero is undergoing an evaluation process: Buchard et al. (2014) evaluated concentrations of SO_2 and SO_4 simulated by GEOS-5 during a field campaign in the United States; Buchard et al. (2015) evaluated MERRAero's assimilation of AOD with many remote sensing instruments over Africa, South America, central and eastern Asia; and Buchard et al. (2016) evaluated surface concentrations of fine PM speciation across the continental U.S. Buchard et al. (2016) obtained a stronger correlation between MERRAero and observations of fine PM concentration from rural networks as opposed to suburban networks. Suburban environments are more likely to contain local sources of pollution which are not captured by MERRAero's resolution. In terms of chemical speciation, overall, the concentrations of different fine PM species were well simulated, although some biases were noticed, such as a large underestimation of OC concentration at suburban locations, especially in winter, and an excessive contribution of DS particles from local and remote sources. The goal of this paper is to expand MERRAero's reliability as a tool to study air quality issues around the world, as well as to identify shortcomings which will help improve subsequent versions, by evaluating the surface concentration of PM over Europe.

2. Methods

MERRAero simulates the concentration of five PM species every hour: SO_4 , OC, BC, DS and SS. Based on these, it is possible to estimate the total concentration of PM_{10} (PM with diameter $\leq 10 \mu\text{m}$) as follows:

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

Brackets denote concentration. $[\text{SO}_4]$ is multiplied by 1.375 since SO_4 is assumed to be fully neutralized by ammonium (NH_4) in the form of ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) and $[\text{OC}]$ is multiplied by 1.8 to account for other organic compounds found in particulate organic matter (POM). Furthermore, since MERRAero differentiates PM sizes of DS and SS, it is possible to estimate $[\text{PM}_{2.5}]$, a widely used air quality indicator, as follows:

$$[\text{PM}_{2.5}] = 1.375 \times [\text{SO}_4] + 1.8 \times [\text{OC}] + [\text{BC}] + [\text{DS}_{2.5}] + [\text{SS}_{2.5}] \quad (2)$$

Eqs. (1) and (2) assume that SO_4 , OC and BC are all in the form of $\text{PM}_{2.5}$ and lack the concentration of nitrate particles, which can make up a considerable portion of total $[\text{PM}_{2.5}]$ (Buchard et al., 2016).

A European network of air quality surveillance stations is running under the European Monitoring and Evaluation Programme (EMEP), whose main goal is to provide to governments with information regarding the concentration of a broad range of air pollutants and their transport across political boundaries (Tørseth et al., 2012). This information is publically available on the EMEP database (<http://ebas.nilu.no>).

We evaluate MERRAero [PM] simulation and its chemical speciation at 55 locations across Europe where measurement data are available between 2003 and 2014 (Fig. 1). According to EMEP guidelines, monitoring stations should be regionally representative and therefore located in rural areas with minimal influence of local pollution sources (Tørseth et al., 2012). A closer look at the locations reveals, however, that a large number of them in Fig. 1 are located in the outskirts of residential settlements, therefore some local sources of PM should be expected. Out of these 55 locations, 52 measure $[\text{PM}_{10}]$ and $[\text{PM}_{2.5}]$ over 24-hour periods, at varying frequencies. Chemical speciation of $[\text{PM}_{2.5}]$ is also available at a number of locations but much less widespread: 32 stations measure $[\text{SO}_4]$, 23 measure the concentration of carbonaceous particles and 24 measure the concentration of fine chloride ions ($\text{Cl}_{2.5}$). $[\text{Cl}_{2.5}]$ is multiplied by 1.8 to obtain $[\text{SS}_{2.5}]$ (Seinfeld and Pandis, 1998). Since few locations measure all necessary minerals to obtain a reasonable estimate of $[\text{DS}_{2.5}]$, this species isn't evaluated. Hourly MERRAero data is averaged on a daily basis and paired at the 55 locations where concentration measurements are available. A spatial consistency algorithm is applied to assure reliability of the observed and simulated concentrations, which goes as follows: since trace concentrations are usually lognormally distributed, the bias between log-simulated concentration and log-observed concentration ($B_{\log} = \log(C_s) - \log(C_o)$; C_s : simulated concentration, C_o : observed concentration) is calculated at all locations on a given day; the average and standard deviation of $B_{\log}$ are calculated and used to define a reliability interval which justifies ~95% of the normal distribution: $B_{\log} \pm 2\sigma_{B_{\log}}$; all data pairs that fall outside this interval are excluded.

Performance statistics are calculated to quantify MERRAero's accuracy: the mean fraction $\bar{F} = \bar{C}_s / \bar{C}_o$, the mean bias $\bar{B} = \bar{C}_s - \bar{C}_o$, the standard deviation of the bias (SD-B) and the correlation coefficient (R); as well as log transformed statistics: $\bar{B}_{\log} = \overline{\log(C_s)} - \overline{\log(C_o)}$, SD- $B_{\log}$ and $R_{\log}$. Despite being widely applied, R is a criticized index for evaluating model performance since it doesn't directly compare simulated with observed data (Willmott, 1982). Therefore, Chang and Hanna (2004) recommended as a rigorous index to evaluate air quality models the proportion of simulated data which falls within a factor of 2 of observed data (FAC2, i.e. proportion of the data which satisfies $0.5 \leq C_o / C_s \leq 2.0$), since this index is not disproportionately sensitive to extreme values and is unaffected by simplification of errors. Chang and Hanna (2004) considered a model's performance to be reasonably good if $\text{FAC2} \geq 0.50$.

3. Results

The spatial consistency algorithm excluded 4.0% of PM_{10} and 4.2% of $\text{PM}_{2.5}$ data pairs. Overall, on average, MERRAero overestimates $[\text{PM}_{10}]$ by a factor of 1.25, which represents an additional $4.1 \mu\text{g}/\text{m}^3$ (Table 1). $[\text{PM}_{2.5}]$, on the other hand, is underestimated by a factor of 1.25 ($2.1 \mu\text{g}/\text{m}^3$). As for chemical speciation, the algorithm excluded 4.5% of SO_4 , 0.7% of OC, 3.2% of BC and 1.0% of $\text{SS}_{2.5}$ data pairs. $[\text{SO}_4]$ and $[\text{BC}]$ are slightly underestimated, by $0.5 \mu\text{g}/\text{m}^3$ and $0.02 \mu\text{g}/\text{m}^3$, respectively. $[\text{OC}]$ is highly underestimated by a factor of 3.18, representing a discrepancy of $2.5 \mu\text{g}/\text{m}^3$, and $[\text{SS}_{2.5}]$ is also overestimated by a large factor of 2.11, but its actual average bias is lower ($1.0 \mu\text{g}/\text{m}^3$).

All species in Table 1, except for OC and $\text{SS}_{2.5}$, have a reasonably low $\bar{B}$ and $\bar{B}_{\log}$, however, there is quite a lot of scattering in the data, as reflected by SD-B and SD- $B_{\log}$ values which are much larger than their respective average B and $B_{\log}$, accompanied by low R and $R_{\log}$ values. It is worth mentioning that SD's are disproportionately influenced by extreme data pairs. For example, if such pairs of data with a factor above 5 are excluded from the sample, which



Fig. 1. Locations of monitoring stations.

Table 1
Performance statistics for the ensemble of locations. AOC stands for “average observed concentration”.

	PM ₁₀	PM _{2.5}	SO ₄	OC	BC	SS _{2.5}
n	133,025	99,191	46,817	13,397	13,165	10,089
AOC (µg/m ³)	16.5	10.5	2.3	3.7	1.0	0.9
F̄	1.25	0.80	0.77	0.31	0.98	2.11
B̄ (µg/m ³)	4.1	−2.1	−0.5	−2.5	−0.0	1.0
SD-B (µg/m ³)	18.0	8.1	1.9	4.7	1.2	3.4
B̄ _{log}	0.18	−0.11	−0.21	−0.84	0.45	1.27
SD-B _{log}	0.72	0.62	0.65	0.93	0.94	1.54
R	0.44	0.47	0.48	0.16	0.52	0.25
R _{log}	0.48	0.59	0.60	0.39	0.60	0.44
FAC2	0.67	0.74	0.70	0.41	0.54	0.29

represents 3.4% of the PM₁₀ and 1.5% of the PM_{2.5} samples, the SD-B's are reduced to 15.2 µg/m³ and 7.0 µg/m³, respectively. Fig. 2 presents density scatter plots which compare observed and simulated concentrations for all species. Although some scatter is clearly noticeable, the plots also show that the bulk of PM₁₀, PM_{2.5} and SO₄ concentration data are reasonably well simulated. The slight underestimations of [PM_{2.5}] and [SO₄] are more noticeable in the log-transformed concentration plots (as well as the large

underestimation of [OC] and large overestimation of [SS_{2.5}]). Furthermore, the same species all have a FAC2 above 50%, supporting a favorable performance. With a FAC2 above 70%, [PM_{2.5}] and [SO₄] are overall very well estimated.

The geographical distribution of B is mapped in Fig. 3. The mean bias for [PM₁₀] is reasonably close to zero across Europe except for a few locations where the overestimation is quite obvious: Cyprus, Ireland, southwest Sweden, western Denmark and the Spanish island of Minorca. [PM_{2.5}] is mostly underestimated in central Europe, particularly in Italy, eastern Austria and eastern Germany. The mean bias for [SO₄] is also close to zero except for the large overestimations observed in southern England and Slovenia. [OC] is underestimated across Europe, particularly in Italy. At the opposite, [BC] is overestimated in most of Europe except, once again, in Italy where it is underestimated. [SS_{2.5}]'s overestimation is observed across Europe, but coastal locations tend to be most affected.

Finally, a seasonal analysis is presented in Fig. 4 which graphs a comparison between observed and simulated concentrations for the six pollutants. [PM₁₀] is overestimated throughout most of the year, while [PM_{2.5}] is well simulated during most of the spring, summer and fall, but underestimated in winter. The underestimation is also larger in winter for [SO₄], [OC] and [BC]. The [OC] underestimation is particularly noticeable from November to

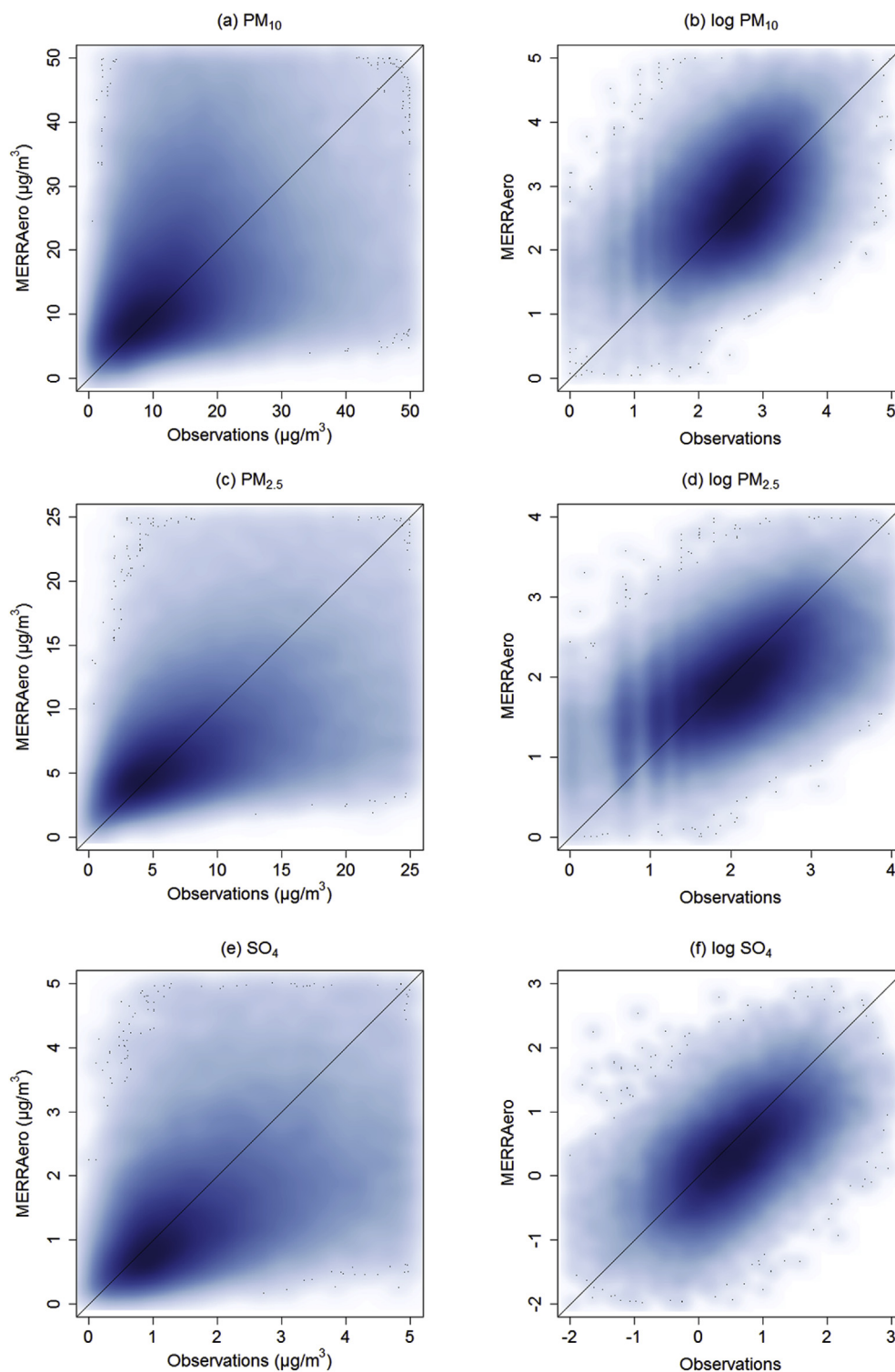


Fig. 2. Density scatter plots between observations and simulations of PM concentrations and species over the ensemble of European locations.

February by factors up to 6.6. $[SS_{2.5}]$ is overestimated throughout the whole year.

4. Discussion

Although the comparisons of $[SO_4]$, $[OC]$, $[BC]$ and $[SS_{2.5}]$ in Fig. 4 are not directly comparable between each other or to those of

$[PM_{2.5}]$ due to different samples and measurement techniques, they provide clues that explain the discrepancy between observed and simulated $[PM_{2.5}]$. Aside from the lack of nitrate particles, the wintertime underestimation of $[PM_{2.5}]$ is likely caused by a combination of $[SO_4]$, $[OC]$ and $[BC]$ underestimations during the same months. This concentration discrepancy for carbonaceous particles is similarly observed in suburban locations in the U.S. (Buchard

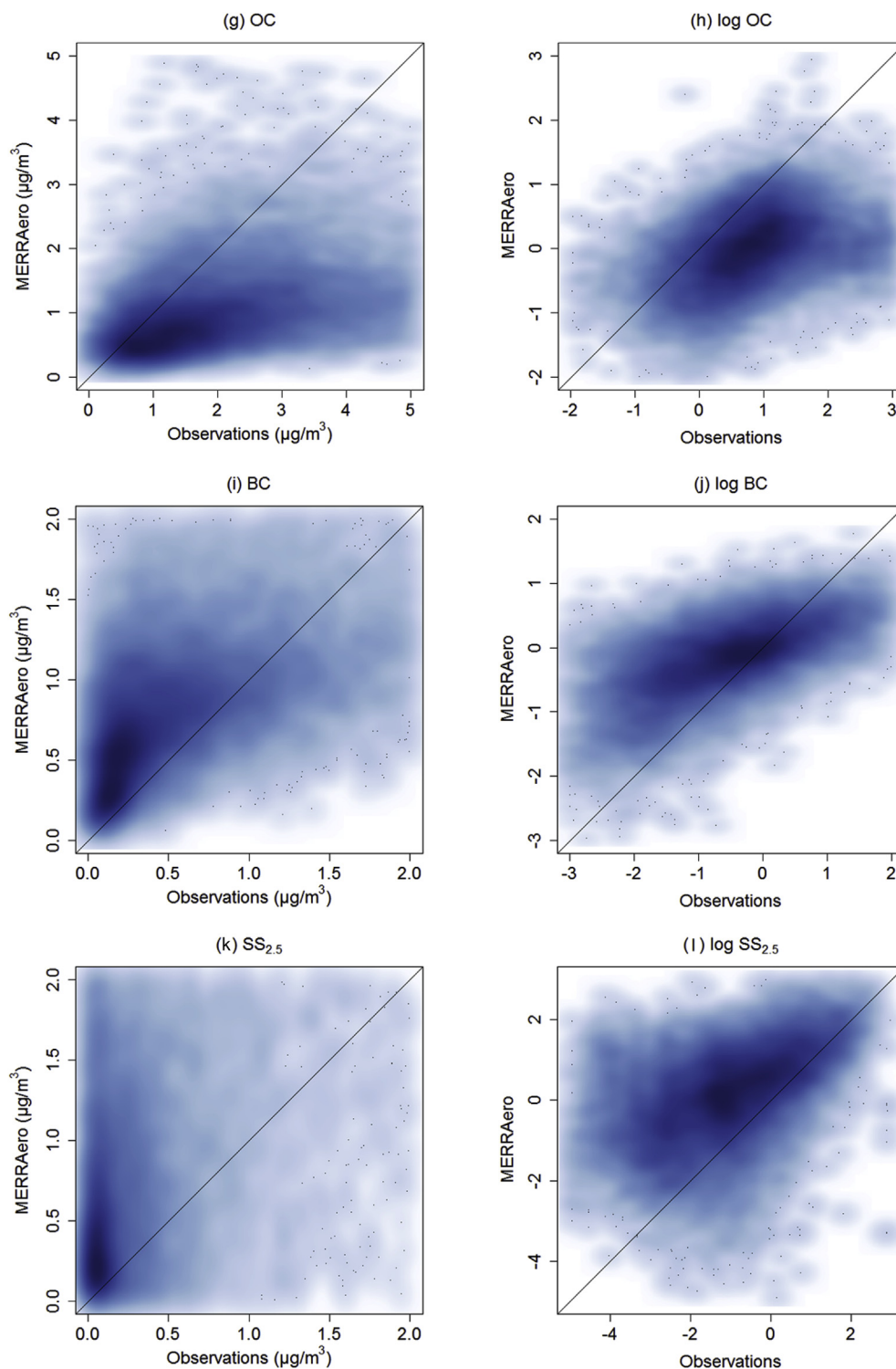


Fig. 2. (continued)

et al., 2016) where it was hypothesised that anthropogenic sources of such particles, such as residential wood burning, unaccounted for in MERRAero's resolution significantly increase a city or a town's PM load in winter (Cooper, 1980; Sexton et al., 1984; Hellén et al., 2008; Pandis et al., 2016). There is indeed a stark contrast between these observations and those in rural U.S. where maximum [OC] and [BC] occur in summer, caused by seasonal wildfires

(Buchard et al., 2016). To a lesser extent, SO_2 is also emitted from biomass burning (Haywood and Boucher, 2000), which could explain the larger discrepancy between observed and simulated wintertime $[\text{SO}_4]$. Performance statistics were recalculated for $[\text{PM}_{2.5}]$, $[\text{SO}_4]$, [OC] and [BC] between April and September only, producing favorable results (Table 2), and, by extension, suggesting a misrepresentation of wintertime anthropogenic sources of PM by

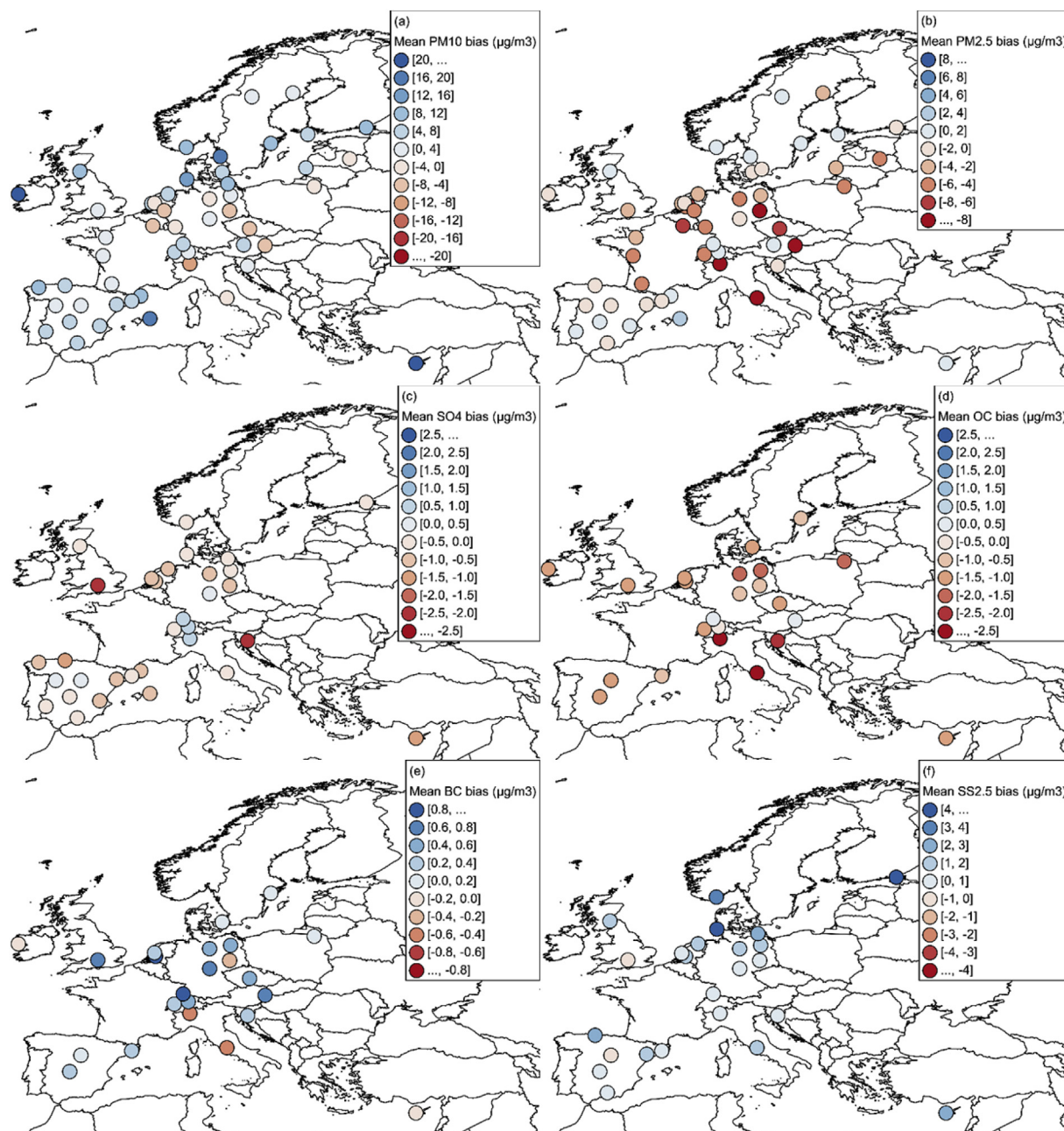


Fig. 3. Maps of the mean bias for PM concentrations and species.

MERRAero. MERRAero reproduces a concentration of SO₄ larger than OC (Table 3), while the reverse is usually observed over Europe (Table 1; Putaud et al., 2004, 2010). [SS_{2.5}] is consistently overestimated by MERRAero, a result also obtained in the U.S. (Buchard et al., 2016), explained in part by measurement biases (Hand et al., 2011). Since coastal locations tend to have the largest $\bar{B}$, this would suggest that SS emissions from the oceans are exaggerated. This overestimation of [SS_{2.5}] slightly compensates for the underestimation of the concentration of other species in simulating [PM_{2.5}].

The shortcomings identified in the previous paragraph regarding MERRAero's simulation of [SO₄], [OC] and [BC] has relatively little impact on the overall simulation of [PM₁₀]. MERRAero's overestimation of [PM₁₀] is rather consistent throughout most of the year compared to its inconsistent underestimation of [PM_{2.5}]

(Fig. 4). Indeed, the simulation of [PM₁₀] is composed by 80% of DS and SS, compared to 47% for [PM_{2.5}] (Table 3). Europe certainly receives a significant amount of DS from the Sahara desert (d'Almeida, 1986), even in the northern region (Ansmann et al., 2003), and SS from the surrounding seas (Tsyro et al., 2011). MERRAero's [PM₁₀] performance therefore relies predominantly on simulating particles originating from natural sources. Unfortunately, given the scarcity of direct coarse particle speciation measurements, we are unable to reliably evaluate this for the time being. If [SS] is overestimated to the same extent as [SS_{2.5}], this, coupled with the large underestimation of [OC], would conceivably reproduce the results obtained in Table 1.

At this point, it is difficult to speculate as to why MERRAero is performing better in some regions of Europe compared to others. A

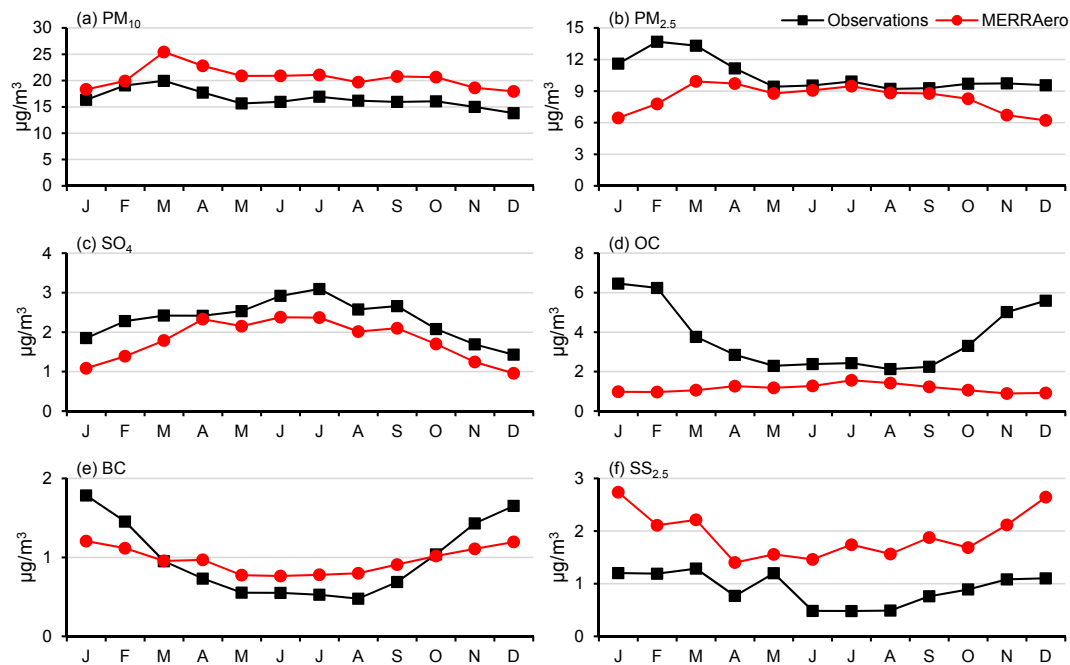


Fig. 4. Monthly averages of PM concentration and species observations and simulations over the ensemble of European locations.

Table 2
Performance statistics for the ensemble of locations between April and September. AOC stands for “average observed concentration”.

	$\text{PM}_{2.5}$	SO_4	OC	BC
n	50,532	23,581	6856	6640
AOC ($\mu\text{g}/\text{m}^3$)	9.7	2.7	2.4	0.6
$\bar{F}$	0.93	0.82	0.56	1.42
$\bar{B}$ ($\mu\text{g}/\text{m}^3$)	−0.6	−0.5	−1.1	0.2
SD-B ($\mu\text{g}/\text{m}^3$)	5.1	1.9	1.6	0.6
$\bar{B}_{\log}$	−0.03	−0.14	−0.51	0.64
SD- $B_{\log}$	0.48	0.62	0.72	0.84
R	0.62	0.47	0.30	0.42
$R_{\log}$	0.66	0.54	0.40	0.54
FAC2	0.85	0.73	0.54	0.54

country where MERRAero is performing particularly poorly is Italy. $[\text{PM}_{2.5}]$, [OC] and [BC] are all well underestimated at both Italian stations. Their respective average concentrations are measured at $21.7 \mu\text{g}/\text{m}^3$, $7.4 \mu\text{g}/\text{m}^3$ and $1.8 \mu\text{g}/\text{m}^3$ in northern Italy, and $20.0 \mu\text{g}/\text{m}^3$, $11.7 \mu\text{g}/\text{m}^3$ and $1.1 \mu\text{g}/\text{m}^3$ in central Italy, substantially higher than almost anywhere else in Europe. The station in northern Italy is located in the vicinity of many small towns, 50 km away from Milan and close to the Alps. Its location is ideal to accumulate clustered pollutants originating from urban sources unaccounted

for by MERRAero. The station in central Italy is also located west of the Apennine Mountains which cluster pollution originating from nearby towns. If those two stations are removed from the sample, the evaluations of $[\text{PM}_{2.5}]$ and [OC] are indeed substantially improved (Table 4). The wintertime bias remains but is much less pronounced (Fig. 5). The fact that MERRAero doesn't simulate secondary organic carbon particles might also contribute to the underestimation.

Table 4
Performance statistics for the ensemble of locations without the Italian stations. AOC stands for “average observed concentration”.

	$\text{PM}_{2.5}$	OC
n	95,514	9737
AOC ($\mu\text{g}/\text{m}^3$)	10.1	2.2
$\bar{F}$	0.82	0.50
$\bar{B}$ ($\mu\text{g}/\text{m}^3$)	−1.9	−1.1
SD-B ($\mu\text{g}/\text{m}^3$)	7.5	1.8
$\bar{B}_{\log}$	−0.10	−0.59
SD- $B_{\log}$	0.61	0.75
R	0.47	0.37
$R_{\log}$	0.58	0.47
FAC2	0.75	0.50

Table 3
Average concentrations simulated by MERRAero for the ensemble of locations in Fig. 1 over the study period.

Species	Average concentration ($\mu\text{g}/\text{m}^3$)	Proportion of PM_{10} (%)	Proportion of $\text{PM}_{2.5}$ (%)
PM_{10}	21.7	—	—
$\text{PM}_{2.5}$	8.5	39.1	—
$(\text{NH}_4)_2\text{SO}_4$	2.3	10.7	27.2
POM	1.8	8.1	20.6
BC	0.7	3.0	7.7
DS	6.8	31.2	—
SS	10.2	47.1	—
$\text{DS}_{2.5}$	2.0	9.0	23.0
$\text{SS}_{2.5}$	1.8	8.4	21.5

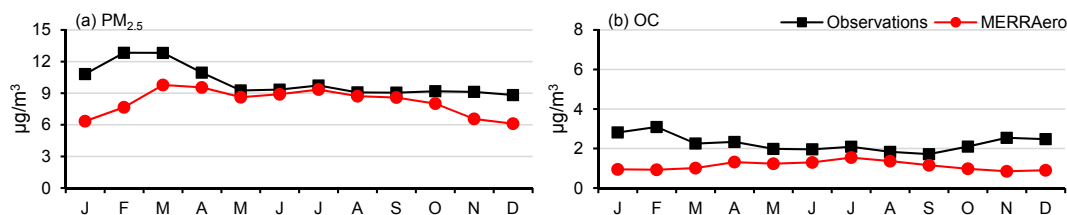


Fig. 5. Monthly averages of PM concentration and species observations and simulations over the ensemble of European locations without the Italian stations.

5. Conclusion

We evaluated MERRAero's simulation of PM₁₀ and PM_{2.5} speciation at the surface over Europe. Overall, MERRAero simulates well [PM₁₀], [PM_{2.5}], [SO₄] and [BC]. Despite considerable scattering within the dataset, the average biases and, most importantly, the FAC2, a rigorous index to evaluate air quality models, reproduced favorable results. The evaluation of [OC] wasn't as encouraging, revealing important underestimations overall, especially in winter. Wintertime underestimations were also obtained in the comparisons of [SO₄] and [BC], and by extension in [PM_{2.5}]. Anthropogenic emissions of carbon and sulfate in the form of residential wood burning, unresolved by MERRAero, are a likely explanation for this discrepancy. The evaluation of [PM_{2.5}], [SO₄] and [OC] did improve during the summer. By removing two stations from the sample where measurements of [OC] were particularly high, the evaluation of this species also improved substantially. MERRAero consistently overestimated [SS_{2.5}], by a factor of 2 on average. The bias was most evident at coastal locations which suggests that oceanic SS emissions are inflated.

This article demonstrated that MERRAero is a reliable tool to simulate the concentrations of major aerosol species in most circumstances. Some improvements are however feasible. The major shortcoming identified pertains to MERRAero's resolution, which hopefully will be improved in the foreseeable future. Inclusion of nitrate particles would also be a significant addition and would improve the simulation of [PM_{2.5}]. The simulation could also benefit from moderated sea salt emissions.

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