Evaluation of $PM_{2.5}$ surface concentration simulated by Version 1 of the NASA's MERRA Aerosol Reanalysis over Israel and Taiwan

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Abstract

Version 1 of the NASA MERRA Aerosol Reanalysis (MERRAero) assimilates bias-corrected aerosol optical depth (AOD) data from MODIS-Terra and MODIS-Aqua, and simulates particulate matter (PM) concentration data to reproduce a consistent database of AOD and PM concentration around the world from 2002 to the end of 2015. The purpose of this paper is to evaluate MERRAero's simulation of fine PM concentration against surface measurements in two regions of the world with relatively high levels of PM concentration but with profoundly different PM composition, those of Israel and Taiwan. Being surrounded by major deserts, Israel's PM load is characterized by a significant contribution of mineral dust, and secondary contributions of sea salt particles, given its proximity to the Mediterranean Sea, and sulfate particles originating from Israel's own urban activities and transported from Europe. Taiwan's PM load is composed primarily of anthropogenic particles (sulfate, nitrate and carbonaceous particles) locally produced or transported from China, with an additional contribution of springtime transport of mineral dust originating from Chinese and Mongolian deserts. The evaluation in Israel produced favorable results with MERRAero slightly overestimating measurements by 6% on average and reproducing an excellent year-to-year and seasonal fluctuation. The evaluation in Taiwan was less favorable with MERRAero underestimating measurements by 42% on average. Two likely reasons explain this discrepancy: emissions of anthropogenic PM and their precursors are largely uncertain in China, and MERRAero doesn't include nitrate particles in its simulation, a pollutant of predominately anthropogenic sources. MERRAero nevertheless simulates well the concentration of fine PM during the summer, when Taiwan is least affected by the advection of pollution from China.

Keywords: MERRAero, Evaluation, Fine particulate matter, Israel, Taiwan.

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INTRODUCTION

NASA's Modern-Era Retrospective Analysis for Research and Application (MERRA, Rienecker *et al.*, 2011) is a reanalysis tool integrating satellite observations from the Earth Observing System and model data from the 5th version of the Goddard Earth Observing System (GEOS-5) atmospheric model and data assimilation system (Rienecker *et al.*, 2008) in order to produce a consistent database in both time and space of various environmental variables around the world since the beginning of the satellite era. Recently, 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 as well as the Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) model (Chin *et al.*, 2002) were included in MERRA to create a reanalysis of aerosols labelled "MERRAero". GOCART simulates the sources, sinks, transport and concentration of sulfate (SO_4), organic carbon (OC), black carbon (BC), dust (DS) and sea salt (SS) aerosols (Chin *et al.*, 2002; Colarco *et al.*, 2010). 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 (SO_2, the precursor of SO_4) 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).

MERRAero simulates the concentration 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) from 2002 to the end of 2015 (Buchard *et al.*, 2015). Considering that these aerosol species, also referred to airborne particulate matter (PM), affect public health and visibility differently

63 depending on their size and chemistry (e.g., Laden *et al.*, 2000; Schwartz and Neas, 2000; Groblicki *et*
64 *al.*, 1981), MERRAero's differentiation of the aerosols' chemical speciation is a significant improvement
65 for studying a broad range of air quality issues around the world since very few monitoring networks
66 make such a distinction of local PM observations, but especially in regions with unreliable or scarce
67 monitoring.

68 Different components of MERRAero have been evaluated in different regions of the world. Its
69 assimilation of AOD has been validated over Africa, South America, central and eastern Asia using many
70 remote sensing instruments (Buchard *et al.*, 2015); in the United States, the surface concentrations of
71 $PM_{2.5}$, their chemical speciation and SO_2 has been thoroughly evaluated (Buchard *et al.*, 2014; 2016);
72 and in Europe, an evaluation of the surface concentrations of PM_{10}, $PM_{2.5}$ and some of their chemical
73 speciation has been performed (Provençal *et al.*, 2016). The concentrations of PM_{10}, $PM_{2.5}$ and SO_4 were
74 generally well simulated in both the U.S. and Europe but Buchard *et al.* (2016) and Provençal *et al.*
75 (2016) noticed an underestimation of carbonaceous concentration in urban/suburban locations,
76 particularly in winter, due to unresolved sources by MERRAero.

77 The U.S. and Europe have similar PM signatures in the sense that both regions are highly
78 industrialized and therefore anthropogenic particles contribute significantly to their PM load. At the same
79 time, implementation of air quality regulation has successfully reduced the emissions of various
80 atmospheric pollutants across the U.S. and Europe over the last decades (e.g. Granier *et al.*, 2011;
81 Klimont *et al.*, 2013; Hand *et al.*, 2012; Xing *et al.*, 2013; de Gouw *et al.*, 2014; Vestreng *et al.*, 2007)
82 and, as a result, maintained relatively low levels of PM concentration. There are nevertheless important
83 differences with respect to the chemical speciation of PM between the two regions such as a
84 predominance of carbonaceous particles over the western U.S. due to summer wildfires and a
85 predominance of dust particles over southern Europe due to its proximity to the Sahara desert.

The PM signature in the U.S. and Europe is not representative of many other regions in the world where PM sources and pollution control are profoundly different. In order for MERRAero to achieve optimal reliability for studying air quality issues around the world, the purpose of this article is to pursue MERRAero's evaluation in regions with different and distinct aerosol signatures, those of Israel and Taiwan. The evaluation in Israel, a region with a heavy PM load due to its proximity to major deserts, will ascertain MERRAero's ability to simulate the concentration of aerosol originating from natural sources. Taiwan being located in a region of the world which is routinely experiencing severe air pollution episodes, the evaluation there will provide insight on MERRAero's applicability in highly polluted regions where its contribution would be most beneficial.

LOCATIONS AND METHODS

Israel and Taiwan

Israel is located in western Asia, surrounded by the Mediterranean Sea, the Sahara desert and the Middle Eastern deserts. Its PM concentration load is relatively high, composed largely of mineral DS (Kushelevsky *et al.*, 1983; Malenky *et al.*, 1983; Foner and Ganor, 1992) with an occasionally important contribution from SS particles when the wind is blowing inland (Foner and Ganor, 1992). PM concentration in urban areas such as the coastal city of Tel Aviv is even higher due to anthropogenic SO_4 locally produced or transported from Europe (Foner and Ganor, 1992). Rural locations in Israel have also been impacted by the advection of SO_4 particles originating from Europe (Luria *et al.*, 1989).

Taiwan is an island located in eastern Asia, separated from mainland China by the Taiwan Strait. Its concentration level of PM is fairly high, especially in urban areas (Chen *et al.*, 1999), caused by industrial and transportation activities within Taiwan but also due to wintertime synoptic features that

transport polluted air from China (Lin *et al.*, 2005). SO_4, OC, BC, nitrate (NO_3) and ammonium (NH_4) particles together compose a large portion of PM concentration (Lin *et al.*, 2008; Lin, 2002; Chen *et al.*, 2003; Tsai and Kuo, 2005; Tsai and Cheng, 1999; 2004). Taiwan nevertheless enjoys cleaner air during the summer, coinciding with the typhoon season which sweeps the island with strong winds and heavy rain (Lin *et al.*, 2008). In spring, Taiwan is also impacted by the advection of dust originating from Chinese and Mongolian deserts (Chen *et al.*, 2004).

Evaluation method

MERRAero simulates the concentration of five $PM_{2.5}$ (PM with diameter ≤ 2.5 μm) species every hour: SO_4, OC, BC, $DS_{2.5}$ and $SS_{2.5}$. From these, it is possible to apply a mass reconstruction method to estimate the total concentration of $PM_{2.5}$. Chow *et al.* (2015) reviewed 11 commonly used equations to reconstruct PM mass from speciation measurements which are usually determined by the measurements available. The equations usually took the following form:

$$\mathrm{PM} = \mathrm{Inorganic\ ions} + \mathrm{Organic\ matter} + \mathrm{BC} + \mathrm{DS} + \mathrm{SS} \quad (1)$$

Inorganic ions include SO_4, NO_3 and NH_4 ions. When NH_4 measurements were lacking, SO_4 and NO_3 were assumed to be fully neutralized by NH_4 in the form of ammonium sulfate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3) by multiplying their respective concentrations by 1.375 and 1.29; $(NH_4)_2SO_4$ being composed of 73% of SO_4 by mass and NH_4NO_3 being similarly composed of 78% of NO_3. The concentration of inorganic ions was ultimately estimated by: $1.375 \times [SO_4] + 1.29 \times [NO_3]$ (brackets denote concentration). $[NH_4NO_3]$ was occasionally omitted altogether when NO_3 measurements were lacking or unreliable (e.g., Malm *et al.*, 1994).

The concentration of particulate organic matter (POM) was estimated through OC measurements multiplied by a coefficient which took into account other organic compounds found in POM but not measured. Commonly and historically, a coefficient of 1.4 was used (Chow *et al.*, 2015; Turpin and Lim, 2001), but Turpin and Lim (2001) argued that such a value is often too low. They recommended a value of 1.6 ± 0.2 for urban carbonaceous particles, 2.1 ± 0.2 for aged (non-urban) particles and a value as high as 2.6 for biomass burning particles.

Taking into consideration the PM species simulated by MERRAero and given that this evaluation is performed in a combination of urban and non-urban locations, the following reconstruction is used:

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

Eq. 2 lacks the concentration of NO_3 particles whose sources are predominantly anthropogenic in nature (Delmas *et al.*, 1997).

MERRAero's simulation at the surface is compared to hourly observations of $[PM_{2.5}]$ measured at 11 locations in Israel between 2003 and 2014, and 13 locations in and around Taiwan between 2005 and 2014 (Fig. 1). A spatial consistency algorithm is applied to assure reliability of the observed and simulated data 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 within each study areas on a given hour; the average and standard deviation of B_{log} are calculated and used to define a reliability interval which justifies ~95% of the normal distribution: $\overline{B_{log}} \pm 2\sigma_{Blog}$; all data pairs that fall outside this interval are excluded.

Performance statistics are calculated to quantify MERRAero's accuracy: the mean fraction $\overline{F} = \overline{C_s} / \overline{C_o}$, the mean bias $\overline{B} = \overline{C_s} - \overline{C_o}$, the standard deviation of the bias (SD-B) and the correlation coefficient (R). Given that trace concentrations are lognormally distributed, it is also relevant to compute log-transformed statistics: $\overline{B_{log}}$, SD-B_{log} and R_{log}. Willmott (1982) criticized the use of R to evaluate model performance since it doesn't directly compare simulated with observed data. 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 FAC2 ≥ 0.5.

RESULTS AND DISCUSSION

Israel

The spatial consistency algorithm excluded 5% of the data in Israel. At 22.5 µg m^{-3}, the $PM_{2.5}$ load in Israel is high (Table 1) compared to Europe (Provençal *et al.*, 2016), rural and suburban U.S. (Buchard *et al.*, 2016). Overall, MERRAero simulates [$PM_{2.5}$] very well in Israel by slightly overestimating its average concentration by 6% or 1.4 µg m^{-3} (Table 1). However, the high SD-B value and modest R suggest significant scatter within the data and a low bias resulting from simplification of errors. On the other hand, it is worth mentioning that SD's are disproportionately impacted by extreme data pairs. For instance, if such data which fall outside a factor of 5 between observed and simulated concentrations, which represent 2.6% of the sample, $\overline{B}$ and SD-B are reduced to 1.0 µg m^{-3} and 19.6 µg m^{-3},

respectively. Furthermore, the density scatter plot of Fig. 2a reveals that although there is some scatter, the bulk of the data is generally well simulated. This is further supported by a high FAC2 value of 76%. The log-transformed data (Table 1; Fig. 2b) support a similar analysis. Additionally, Fig. 3 compares the annual and monthly fluctuations between simulated and observed data, and illustrates an excellent identity between both datasets.

MERRAero's ability to accurately estimate [$PM_{2.5}$] in Israel relies predominantly on its ability to simulate [$DS_{2.5}$] since [$PM_{2.5}$] is largely composed of this species (Table 2) and, to a lesser extent, its ability to simulate [SO_4] and [$SS_{2.5}$]. The evaluation of [$DS_{2.5}$] in the U.S. revealed important seasonal biases without much impact on the evaluation of [$PM_{2.5}$] given its small contribution to [$PM_{2.5}$] there (Buchard *et al.*, 2016). While the U.S. is mostly impacted by long range transport of DS, this evaluation in Israel would suggest that MERRAero performs well in simulating [$DS_{2.5}$] originating from local sources. [SO_4] has been shown to be well simulated in the U.S. and in Europe (Buchard *et al.*, 2016; Provençal *et al.*, 2016), we therefore have no reason to believe otherwise in this region. MERRAero largely overestimated [$SS_{2.5}$] in both the U.S. and Europe due in part to measurement biases which could very well be the case in Israel. An overestimation of [$SS_{2.5}$] could compensate the lack of nitrate particles in the simulation. In any case, the lack of nitrate particles is likely a minor shortcoming given that they are less abundant than SO_4 and probably contribute little to [$PM_{2.5}$].

Taiwan

The spatial consistency algorithm excluded 3% of the data in Taiwan. The $PM_{2.5}$ load in Taiwan is higher than in Israel (29.8 µg m^{-3}; Table 1). Despite a FAC2 of 59%, MERRAero's performance in Taiwan is much less encouraging. On average, MERRAero underestimates total [$PM_{2.5}$] by 8.8 µg m^{-3}, a factor of 1.42. The SD-B is also high and R is positive but low. Fig. 4a–b reveals that the bulk of the

199 simulated data, 66% to be precise, is indeed underestimated.

200 MERRAero's simulation of [$PM_{2.5}$] in Taiwan is mostly anthropogenic in nature (Table 2) with a
201 significant proportion attributed to SO_4. This information coupled with Fig. 5 which compares annually
202 and monthly averaged simulated and observed concentration reveals a few clues as to why MERRAero's
203 performance is less favorable in Taiwan. The evaluation performs well during the summer (typhoon)
204 season but deteriorates during the rest of the year when Taiwan is most impacted by the advection of
205 pollution from China. The use of a constant inventory of SO_2 emissions from 2005 is problematic in the
206 long term since it is increasingly becoming antiquated with every passing year. Indeed, in 2005, China
207 successfully implemented comprehensive policies to reduce SO_2 emissions. As a result, SO_2 emissions
208 and, by extension, SO_4 concentrations have been decreasing since 2006 (Lu *et al.*, 2010; 2011; Wang
209 and Hao, 2012; Zhang *et al.*, 2012; Klimont *et al.*, 2013; B. Zhao *et al.*, 2013; Y. Zhao *et al.*, 2013). This
210 is reflected in Fig. 5 with a near constant decrease of [$PM_{2.5}$] observations as opposed to the near constant
211 year to year concentrations simulated by MERRAero. SO_2 emission estimates from China are also
212 crippled with uncertainties (Smith *et al.*, 2011). The lack of nitrate particles in the simulation is much
213 more troublesome in Taiwan given that [$PM_{2.5}$] is mostly composed of anthropogenic particles. This
214 would certainly explain a significant portion of the underestimation. Another possible explanation for
215 the wintertime discrepancy, one that's also been highlighted by Provençal *et al.* (2016) for the evaluation
216 in Europe, is local sources of pollution unresolved by MERRAero. While MERRAero's simulation takes
217 into account urban sources of pollution, its resolution is too coarse to capture the urban core of cities.
218 Some monitoring stations in Taiwan (Fig. 1) are located in or around large cities, but none of them are
219 located in their downtown core. We therefore don't expect them to be overly influenced by local sources
220 of pollution. Nevertheless, some influence of unresolved sources should be anticipated. The springtime

maximum observed in Fig. 5 is the likely contribution of long range transport of DS, well captured by MERRAero.

CONCLUSION

We evaluated version 1 of the MERRA Aerosol Reanalysis' ability to simulate the concentration of $PM_{2.5}$ in two regions of the world with relatively high levels of PM concentration but with profoundly different PM composition. Israel is characterized by a high concentration of $PM_{2.5}$ due to its proximity to major deserts and to the highly saline Mediterranean Sea. Its $PM_{2.5}$ load is composed mostly of natural particles (mineral dust and sea salt) with some contribution of anthropogenic particles (sulfate) originating from Israel's urban activities and advection from Europe. Taiwan's high $PM_{2.5}$ concentration is mostly anthropogenic in nature due to Taiwan's own industrial activities and to advection of polluted air from China with some contribution of dust particles originating from east Asian deserts.

The evaluation reproduced favorable results in Israel where MERRAero slightly overestimated actual $PM_{2.5}$ concentration by 6% on average. Although there is scatter within the distribution, most of the simulation is reasonably accurate with over 75% of the simulated data falling within a factor of 2 of measurements. Given that most of $PM_{2.5}$ in Israel is mineral dust, this evaluation supports the assumption that MERRAero performs well in simulating the concentration of fine dust originating from local and regional sources throughout the year.

The evaluation is not as favorable in Taiwan where MERRAero significantly underestimated measured $PM_{2.5}$ concentration by 42% on average. Given that $PM_{2.5}$ in Taiwan is mostly composed of anthropogenic particles, many of which originate from China, two likely reasons explain this outcome: the uncertainty with respect to Chinese emissions and the lack of nitrate particles in the simulation. The

simulation was indeed better during the summer when Taiwan is least impacted by advection of polluted air from China.

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366 **Table 1.** Performance statistics for the ensemble of locations in Israel and Taiwan. AOC stands for

367 "average observed concentration".

	Israel	Taiwan
n	1,016,778	1,024,992
AOC (μg m^{-3})	22.5	29.8
F	1.06	0.70
$\overline{B}$ (μg m^{-3})	1.4	–8.8
SD-B (μg m^{-3})	23.8	22.7
$\overline{B_{log}}$	0.07	–0.27
SD-B_{log}	0.66	0.83
R	0.56	0.27
R_{log}	0.50	0.30
FAC2	0.76	0.59

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Table 2. Average concentration simulated by MERRAero for the ensemble of locations in Fig. 1 over the study period.

Species	Israel		Taiwan	
	Average concentration ($\mu g\ m^{-3}$)	Proportion of $PM_{2.5}$ concentration (%)	Average concentration ($\mu g\ m^{-3}$)	Proportion of $PM_{2.5}$ concentration (%)
$PM_{2.5}$	24.0	–	20.9	–
$(NH_4)_2SO_4$	4.5	18.7	9.3	44.3
POM	1.3	5.4	3.0	14.1
BC	0.4	1.7	0.7	3.4
$DS_{2.5}$	13.9	57.9	2.0	9.3
$SS_{2.5}$	3.9	16.3	6.1	28.9

Table and figure captions

Table 1. Performance statistics for the ensemble of locations in Israel and Taiwan. AOC stands for "average observed concentration".

Table 2. Average concentration simulated by MERRAero for the ensemble of locations in Fig. 1 over the study period.

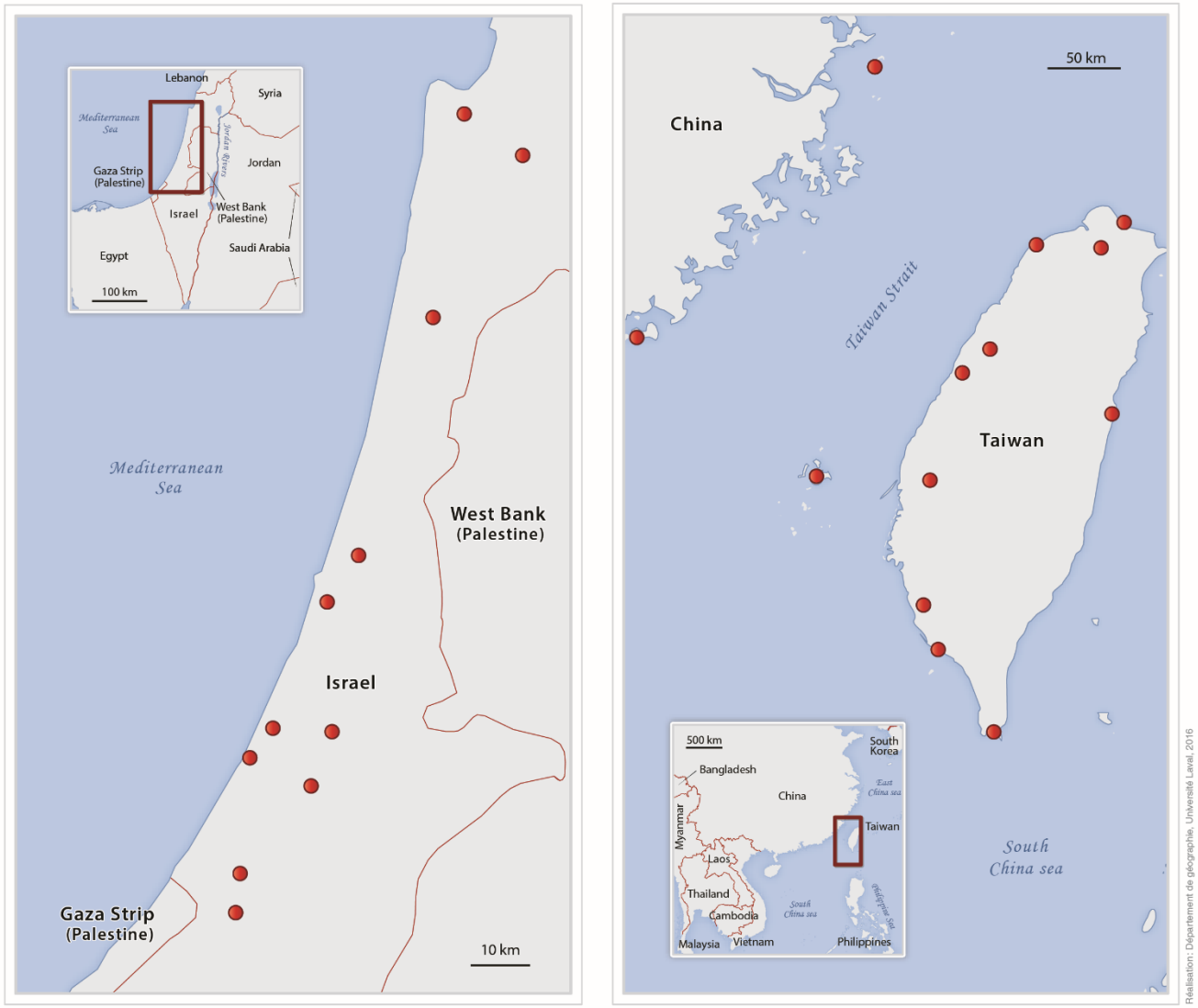
Fig. 1. Location of monitoring stations.

Fig. 2. Density scatter plot for (a) observed and simulated [$PM_{2.5}$], and (b) log transformed observed and simulated [$PM_{2.5}$] for the ensemble of locations in Israel.

Fig. 3. (a) Yearly and (b) monthly average of [$PM_{2.5}$] observation, simulation and bias, and (c–d) similarly for the SD and FAC2, for the ensemble of locations in Israel.

Fig. 4. Density scatter plot for (a) observed and simulated [$PM_{2.5}$], and (b) log transformed observed and simulated [$PM_{2.5}$] for the ensemble of locations in Taiwan.

Fig. 5. (a) Yearly and (b) monthly average of [$PM_{2.5}$] observation, simulation and bias, and (c–d) similarly for the SD and FAC2, for the ensemble of locations in Taiwan.



390

391 **Fig. 1.** Location of monitoring stations.

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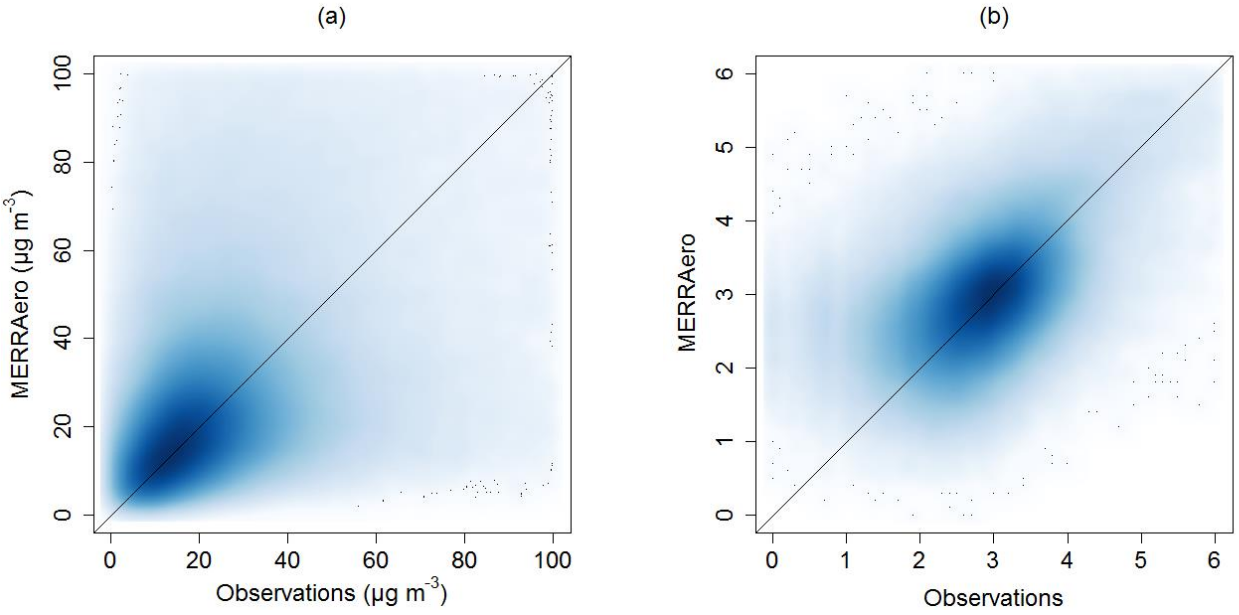

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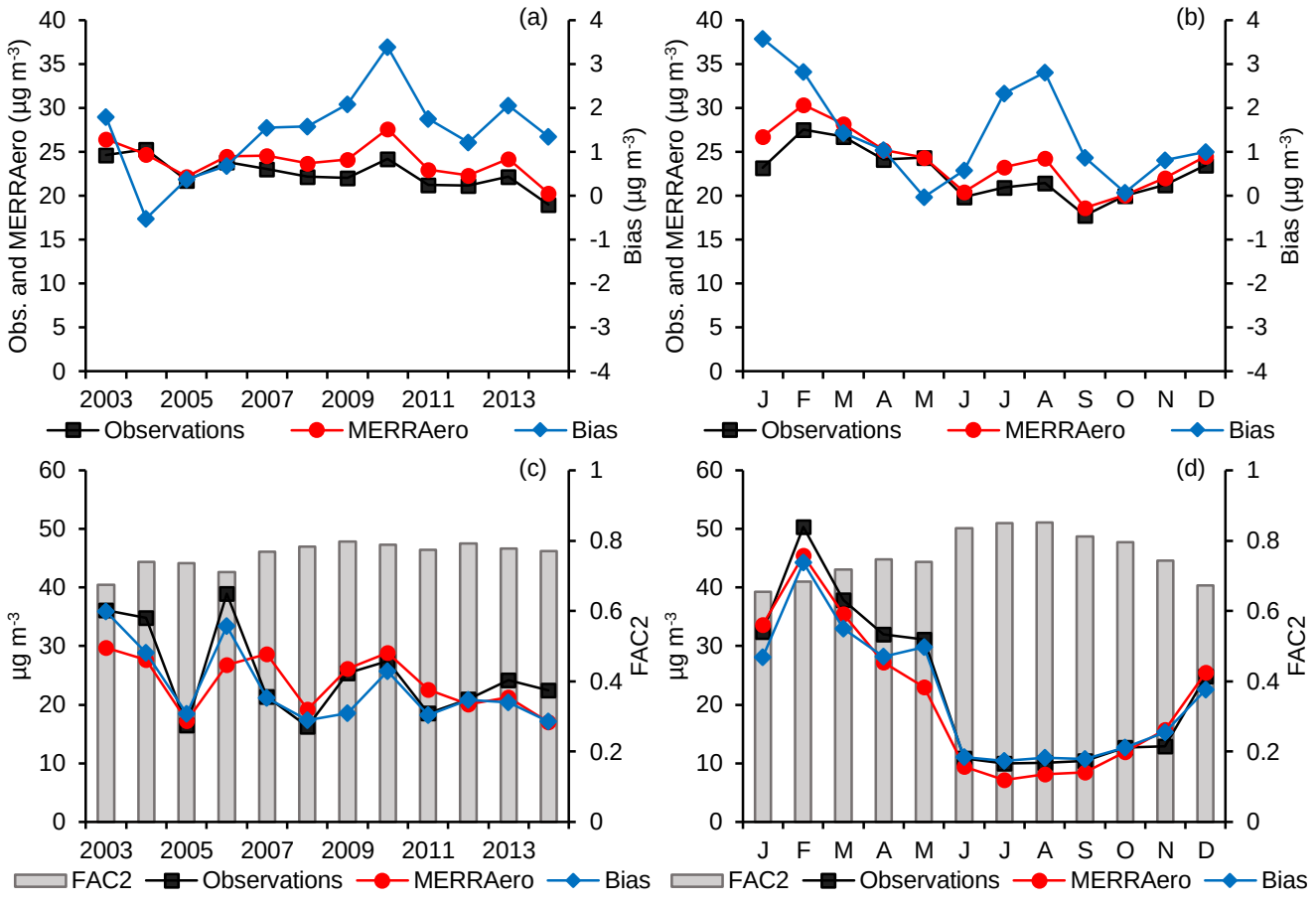

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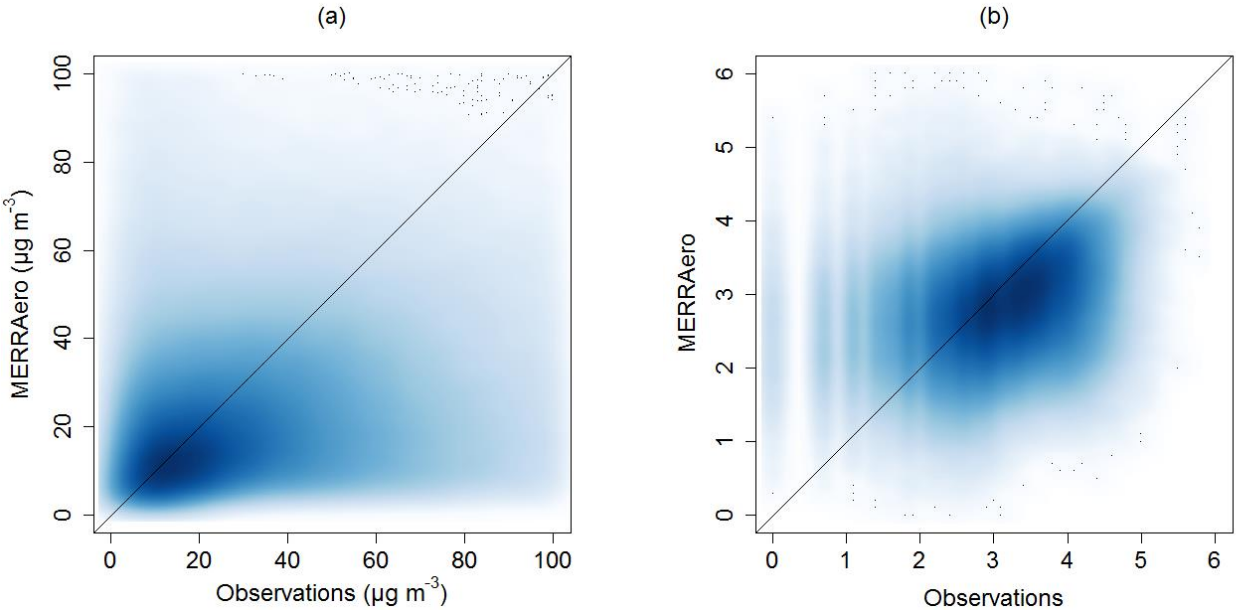

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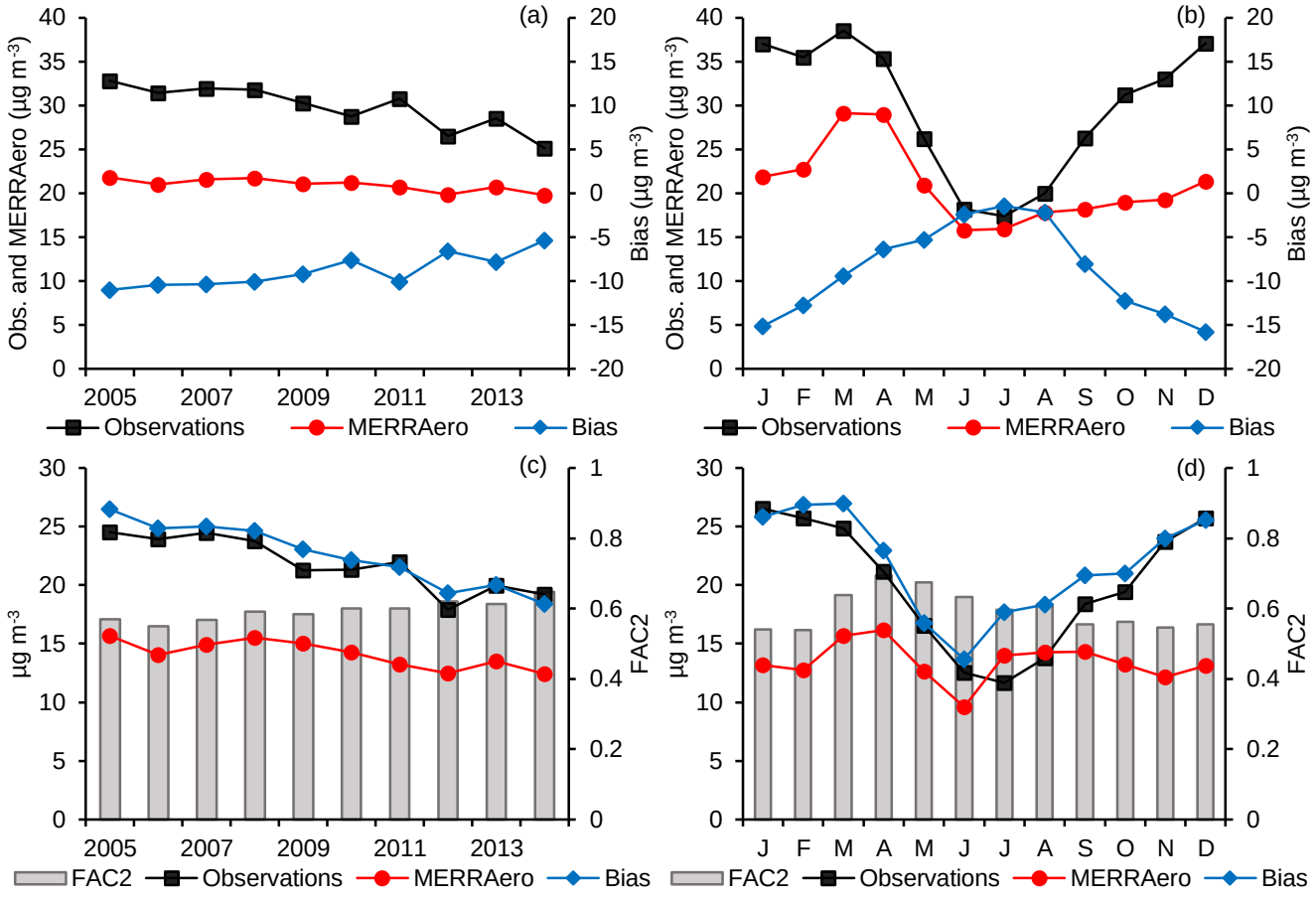

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