Current State of the global operational aerosol multi-model ensemble: an update from the International Cooperative for Aerosol Prediction (ICAP)

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Abstract

Since the first International Cooperative for Aerosol Prediction (ICAP) multi-model ensemble (MME) study, the number of ICAP global operational aerosol models has increased from five to nine. An update of the current ICAP status is provided, along with an evaluation of the performance of the ICAP-MME over 2012-2017, with a focus on the June 2016-May 2017 time period. Evaluated with ground based Aerosol Robotic Network (AERONET) aerosol optical depth (AOD) and data assimilation quality Moderate Resolution Imaging Spectroradiometer (MODIS) retrieval products, the ICAP-MME AOD consensus remains the overall top scoring and most consistent performer among all models in terms of root mean square error (RMSE), bias and correlation for total, fine and coarse mode AODs as well as dust AOD; this is similar to the first ICAP-MME study. Further, over the years, the performance of ICAP-MME is relatively stable and reliable compared to more variability in the individual models. The extent to which the AOD forecast error of the ICAP-MME can be predicted is also examined. Leading predictors are found to be the consensus mean and spread. Regression models of absolute forecast errors

were built for AOD forecasts of different lengths for potential applications. ICAP-MME performance in terms of modal AOD RMSEs of the 21 regionally representative sites over 2012-2017 suggests a general tendency for model improvements in fine-mode AOD, especially over Asia. No significant improvement in coarse-mode AOD is found overall for this time period.

Key words: aerosol, ensemble, aerosol forecast, multi model ensemble, global aerosol model, probabilistic forecast, operational aerosol forecast, aerosol modeling

1. Introduction

Over the past decade, global aerosol modeling has grown from a largely climate and geophysical science activity to include operational forecasting and decision support systems. Weather, air quality, and health communities are increasingly relying on aerosol analysis and forecast products. For example, near-real-time (NRT) aerosol forecasts are used to provide situational awareness for civilian aviation, military operations, and air-quality alerts. Operationally, aerosol particles can also interfere with many aspects of modern-day Earth system observing systems, including retrievals of sea surface temperature (e.g., May et al., 1992, Reynolds, 1989, Bogdanoff et al., 2015), ocean color (e.g., Gordon, 1997), and land use systems (Song et al. 2001), as well as atmospheric retrievals of temperature, water vapor and other gases, which are used to constrain atmospheric states in numerical weather prediction (NWP) models (Houweling, et al., 2005). Indeed, progress has been made in accounting for aerosol impacts on radiances in satellite retrievals through atmospheric corrections (Weaver et al., 2007; Wang and Niu, 2013) and aerosol direct and indirect impact on NWP forecasts (e.g., Mulcahy et al., 2014; Toll et al., 2016).

With the rapid increase in the number of operational and quasi-operational global aerosol models, the International Cooperative for Aerosol Prediction (ICAP) was founded in 2010 (Reid et al., 2011; Benedetti et al., 2011; Colarco et al., 2014a) with one of its goals being the development of a global multi-model aerosol forecasting ensemble (ICAP-MME) for basic research and eventual operational use. The ICAP community, which consists of developers from forecasting centers and remote-sensing data providers, has met yearly since its inception to discuss issues pertaining to operational aerosol forecasting with topics ranging from aerosol

observability (Reid et al., 2011), model validation and verification (Benedetti et al. 2011), aerosol processes, and aerosol data assimilation (http://icap.atmos.und.edu/).

As a relatively new community compared to NWP, ICAP has positioned itself to take advantage of best practices from the NWP community, including methodologies for data assimilation, single-model ensembles (Molteni et al., 1996; Toth and Kalnay, 1997), multi-model ensembles (Park et al., 2008), and consensus products (Sampson et al., 2008). In particular, the motivation for developing the ICAP global multi-model ensemble AOD consensus is based on NWP studies that have shown the usefulness of ensemble-based predictions in understanding systematic errors that arise from the imperfect nature of models and the sensitivity of models to initial conditions. For example, multi-model consensuses are found on average to produce more accurate forecasts of cyclone track and intensity than the individual model members (e.g., Goerss, 2000; Sampson et al., 2008). Likewise, the ICAP-MME aerosol forecast consensus generally performed better than the individual models in the first ICAP-MME global assessment (Sessions et al., 2015).

The first ICAP-MME, as described in Sessions et al. (2015), included four complete aerosol forecast models (European Center for Medium-Range Weather Forecasts-Monitoring Atmospheric Composition and Climate Model (ECMWF-MACC), now under the Copernicus Atmosphere Monitoring Service (CAMS); Fleet Numerical Meteorology and Oceanography Center (FNMOC)/Naval Research Laboratory (NRL)-Navy Aerosol Analysis and Prediction System (NAAPS); Japan Meteorological Agency (JMA)-Model of Aerosol Species in the Global Atmosphere (MASINGAR); and NASA Global Modeling and Assimilation Office (GMAO) Goddard Earth Observing System, Version 5 (GEOS-5); and one dust-only model, (NOAA NCEP NOAA Environmental Modeling System (NEMS) Global Forecast System (GFS) Aerosol Component (NGAC)). Since then, forecast contributions from other forecasting centers have

been added to the ICAP-MME, including dust aerosol forecasts from the Barcelona Supercomputing Center (BSC) and the UK Met Office (UKMO) dust models, and full-species aerosol forecasts from Météo France Modèle de Chimie Atmospherique à Grande Echelle (MOCAGE) and Finnish Meteorological Institute (FMI) System for Integrated modeling of Atmospheric coMposition (SILAM). Additionally, numerous updates were made to improve the quality of predictions from the individual forecast models (e.g. better representation of aerosol processes, more aerosol species, finer spatial and temporal resolution, from offline to inline modeling, etc.), the initial conditions (e.g. new observation types, improved methods for processing and screening observations, and improved data assimilation techniques), and improvements in the driving meteorological model data. As a result of these many updates, a new performance evaluation of the ICAP-MME was deemed necessary for the aerosol forecasting community, as well as joining the larger ensemble community in celebrating 25-years of ensemble prediction at ECMWF (Buizza and Richardson, 2017).

The individual aerosol models that contribute to the ICAP-MME are independent in their underlying meteorology and often in their aerosol sources, sinks, microphysics and chemistry. The diversity of aerosol representation across the aerosol forecast models, similar to that found in aerosol climate models (Kinne et al., 2006), results in differences in predicted aerosol properties and spatial/temporal distributions. In order to increase the accuracy of aerosol forecasts, several centers have employed data assimilation of satellite-and/or-ground-based observations of aerosol optical depth (AOD)-the most widely available and evaluated aerosol parameter. For these models, the diversity in assimilation methods, and the assimilated AOD observations, including the treatments of the observations prior to assimilation (quality control, bias correction, aggregation and sampling, etc.), also leads to differences in the AOD analyses

and forecasts. In this paper, AOD at 550nm from all models are evaluated regionally by representative Aerosol Robotic Network (AERONET) sites and globally using a data assimilation grade satellite aerosol product. We present the basic verification characteristics of the ICAP-MME and their evolution with time, and identify regions of diversity in model analyses and forecasts across the ensemble members. We also evaluate the usefulness of ICAP model ensemble mean and spread for absolute forecast error estimate. Finally, we use this knowledge to build forecast error regression models for potential applications towards probabilistic forecasts.

2. Methodology

In this section, a brief description is provided of the models that are included in the ICAP-MME and an outline is given of the fundamental metrics for model performance in AOD prediction. Drawing from the members of the ICAP-MME's latest generation of quasi-operational aerosol models, AOD analyses and four day AOD forecasts are analyzed from four multi-species models with AOD data assimilation (core members), ECMWF/CAMS, JMA, NASA GSFC/GMAO, and NRL/FNMOC. For the evaluation of dust analyses and forecasts, the UKMO dust model with dust AOD assimilation is included. For dust forecast evaluation only, dust products are included from NOAA NGAC and the BSC CTM, which exclude data assimilation, leading to a total of 7 dust models. As per the ICAP agreement, individual models and their associated metrics are not specified. Instead, the metrics are provided for the ensemble as a whole, as was done in the first ICAP-MME paper (Sessions et al., 2015) with an emphasis on the relative spread of performance for both analyses and forecasts at different sites and regions. The main analysis is conducted over

a one year time period, June 2016 to May 2017, when the most recent validation data from both AERONET and DA-quality satellite products is most abundant. Additionally, the start date of the analysis time, June 2016, coincides with the operational transition of AOD data assimilation for one of the core members, giving a total of four multi-species aerosol models with data assimilation for evaluation.

2.1 Input models

The current ICAP-MME operation includes seven comprehensive global aerosol models: the ECMWF Copernicus Atmosphere Monitoring Service (CAMS, former MACC), GEOS-5, NAAPS, MASINGAR, NGAC, MOCAGE, SILAM, and two dust only global models: BSC Multiscale Online Nonhydrostatic AtmospheReCHemistry model (MONARCH, former BSC-CTM) and UKMO Unified Model. The basic properties and configurations of these participating models are outlined in Table 1 and detailed descriptions of individual models are given in Appendix A. During the study period, there were insufficient data to fully evaluate the MOCAGE, SILAM, and NGAC full-species models with the exception of the NGAC dust component. Therefore, while descriptions are included of these models, MOCAGE, SILAM and the non-dust species of NGAC are not used in the evaluation presented here.

The ICAP models are mostly driven by independent operational/quasi-operational meteorological models that are developed at each NWP/research center, and aerosol variables are either calculated dynamically and concurrently with the meteorological fields ("inline") or run in a separate calculation forced by stored NWP fields ("offline"). Depending on the resolution of the underlying meteorology, the aerosol models have different horizontal and vertical resolutions, ranging from 0.25°x0.31° latitude/longitude and 72 vertical layers to 1.4°x1° and 24

layers. All of the models include dust aerosol, although with different size bins. The comprehensive models carry a full set of aerosol species, including dust, sea salt, biomass burning smoke (combined black carbon and organic carbon from some models) and varying forms of pollution aerosols (sulfate and possible nitrates). The aerosol sources (e.g., biomass burning emissions), sinks, microphysics, and chemistry are also quite different across the models, with the exception of NOAA NGAC and GEOS-5 which use a similar aerosol module.

For the models that have aerosol data assimilation, aerosol forecasts are initialized with analysis fields from their respective DA systems, ranging from 2-dimensional variational (2DVar), 3DVar, and 4DVar to ensemble systems. One consistency across these data assimilation systems is the use of data from the Moderate Resolution Imaging Spectroradiometer (MODIS) with its daily global spatial coverage. However, the treatments applied to the MODIS observations are different among the members. For example, FNMOC/NRL applies strict quality assurance and quality control processes to convert MODIS level 2 data into filtered, corrected and aggregated AOD observations with associated uncertainty estimates. This processing is described for MODIS Collection 5 over-ocean Dark Target AOD by Shi et al. (2011), and over-land by Hyer et al. (2011). For MODIS Collection 6 data, all correction and filtering coefficients were recalculated, and the method used to screen and correct Dark Target over-land retrievals was applied to Deep Blue retrievals also. NASA GMAO also uses MODIS, but adapts a neural network retrieval trained using AERONET data to translate observed MODIS radiances into ground-based calibrated AOD (Randles et al., 2017). The UK Met Office develops their own dust AOD product derived from MODIS retrieved aerosol properties (Pradhan, 2017). Furthermore, since satellite retrieved AOD is a column integrated observation, aerosol speciation and vertical distribution are not constrained by assimilation of AOD. The operations used to

convert AOD into 3-D speciated aerosol fields and vice versa constitute another layer of diversity across models that include data assimilation.

2.2 ICAP-MME

The ICAP-MME is a consensus-style multi-model ensemble where all members are equally weighted. The ensemble of model AODs is generated daily with 1°x1° resolution at 00Z for 6-hourly forecasts out to 120 hours with a 1-day latency. The l-day latency allows aerosol forecasts from all centers, including centers that generate their aerosol predictions on a delayed cycle, to be collected and aggregated into the ICAP-MME. Daily products include AOD distribution maps, mean-spread plots, verification plots, and threat scores as well as the ICAP-MME data itself. Currently, the ensemble is limited to speciated AOD at a standard 550 nm wavelength. It is anticipated, by the end of 2018, that surface mass concentrations of speciated aerosols will also be included in the ICAP-MME. Due to differences in data policy for participating members, plots and data products of each individual member are only available to participating centers. However, plots of ICAP-MME consensus and spread are available on the NRL webpage (http://www.nrlmry.navy.mil/aerosol/) and NetCDF data files including the 550 nm dust, fine-mode (mostly from pollution and biomass burning smoke aerosol), coarse-mode (mostly from dust and sea salt aerosols) and total AOD (from all aerosol species) are available on the US Global Oceans Data Assimilation Experiment (GODAE) website at http://usgodae.org/cgi-bin/datalist.pl?dset=nrl_icap_mme&summary=Go, both last accessed on 31 October 2018.

The data stream for the individual ICAP models and the MME over time is shown in Fig. 1. Because of the operational nature of these models, data stream outages sometimes occur due to

network issues or hardware/software issues, for example. The ICAP-MME is generated daily for the previous 3 days to minimize outages. Most members have data availability greater than 90%, with 6 out of 8 members greater than 95% and the ICAP-MME data is produced with 99.8% availability.

The ICAP MME evaluation results presented in this paper are based on the unweighted arithmetic mean of the ensemble members ("ensemble mean"), and the standard deviation of the ensemble members ("ensemble spread"). We also tested using the ensemble median for the June 2016 to May 2017 study period and our evaluation results and conclusions were unchanged.

   2.3 Verification

The AErosol RObotic NETwork (AERONET, http://aeronet.gsfc.nasa.gov) is a ground-based and global-scale sun photometer network, which has been providing high-accuracy measurements of aerosol properties since the 1990s (Holben et al., 1998). AERONET instruments measure sun and sky radiance at several wavelengths, ranging from the near ultraviolet to near infrared during daytime. It is often used as the primary standard for validating satellite products and model simulations (e.g., Levy et al., 2010; Colarco et al., 2010). For this study, we use the quality-assured AERONET Version 3 Level-1.5 product, which has better cloud-screening and better preservation of high AOD values that were often discarded in previous versions. The complete set of Version 3 cloud screening and quality assurance algorithms and comparisons of the Version 3 product to Version 2 are provided in Eck et al. (2018). While final quality assured Version 3 level 2 data is preferable, complete datasets are posted with delays as long as 18 months, after instruments have been brought back from the field for laboratory recalibration. However, as data is converted to level 2, calibration constants are

back applied to level 1.5 data ensuring the best possible available data while completing timeliness requirements. The Version 3 AERONET data from sites with post-deployment calibration re-processing have AOD uncertainty very similar to AERONET Version 2 Level 2 of ~0.01 in the visible and near-infrared (Eck et al., 1999), since calibration is the dominant source of the measured AOD uncertainty. Other data with only pre-deployment calibration applied (since these instruments are still operating in the field) may have AOD uncertainties that are somewhat higher depending on the magnitude of calibration drift, but will typically be ~0.02 or less since Level 1.5 data have passed the Version 3 cloud-screening and QA filtering. The ICAP DA models have a capability of assimilating AERONET AOD in their research mode (e.g., Rubin et al., 2017), but none of the operational runs apply AERONET AOD assimilation. So the AERONET data serves as an independent dataset for validation purpose.

For this analysis, 21 AERONET sites are selected (Table 2) based on regional representativeness, and the availability of contiguous data records covering June 2016 to May 2017 , the main study period, as well as the longer 2012-2017 period to allow for evaluation of model performance over time. Additionally, sites were selected to maintain as much consistency as possible with the original ICAP-MME evaluation (Sessions et al., 2015), to enable comparisons between previous and current evaluations. Of the original 21 sites, 18 were retained, including 10 sites dominated by dust influence. The 3 sites that were replaced for this analysis due to large data gaps or site decommissioning include the remote oceanic site Crozet Island, which is replaced with Amsterdam Island, the Arabian Peninsula dust site Solar Village replaced with Mezaira, and the South Korea site Baengnyeong replaced with Yonsei University site following the regional representativeness requirement.

Since AERONET instruments do not directly measure at 550nm, measurements from multiple wavelengths (380nm to 1020nm) were used to estimate both fine and coarse mode AODs at 550nm, based on the Spectral Deconvolution Method (SDA) of O'Neill et al. (2001, 2003). The SDA product is capable of capturing the full modal characteristics of fine and coarse particles, based on verifications using in situ measurements (Kaku et al., 2014). SDA derived fine and coarse mode AERONET AODs are then compared to model-predicted fine mode, represented as pollution plus biomass-burning smoke, and coarse mode, represented as sea salt plus dust. To facilitate comparison between ground-based AERONET observations and gridded model output, the 1°x1° degree ICAP model grids in which the AERONET V3 Level 1.5 data fall within are first identified and model AOD is sampled from the identified grid. To account for temporal differences, AERONET data are binned into 6-hour intervals centered at the model synoptic output times of 00, 06, 12 and 18 UTC and then averaged within the bins. AERONET coarse-mode AOD is used for dust AOD validations at sites dominated by dust influence (Table 3) for all models. Fine and total AOD validations only apply to the full-species models.

While AERONET serves as a useful verification dataset due to the small measurement error, sites are only present over land, and are sparse in many regions, limiting the evaluation of global model output. In order to generate a global-scale assessment of model performance, the ICAP-MME analyses and forecasts are also evaluated against the data assimilation quality MODIS C6 AOD product. The methodology to develop the DA-quality MODIS C6 AOD product is similar to that of MODIS C5 AOD product (Shi et al., 2011; Hyer et al., 2011), however, the DA-quality C6 product includes combined AOD retrievals from the Dark-Target and Deep-Blue algorithms, providing more data coverage (compared to Dark-Target only) over desert/bright surfaces (screened using the same methods as the Dark Target over-land retrievals). This DA-quality

MODIS C6 AOD data is a Level 3 product that is produced at the same spatial and temporal resolution as the ICAP-MME products (1°x1^{o} spatial/6-hr temporal resolution). Since the C6 data product includes total AOD only, not speciated or size-resolved AODs, verification against this product is limited to the four full-species aerosol models.

The DA-quality MODIS C6 AOD was derived from MODIS products, which all the ICAP DA model's assimilation systems use to various extents. Thus the verification here is not fully independent. The MODIS C6 AOD used as a verification dataset is not identical to the data assimilated in real time by any of the ICAP models. The MODIS C6 product used for verification was not widely used by ICAP models until early 2017, so what the DA models assimilated during the study period (July 2016-June 2017) were mostly based on the MODIS C5 products. Second, as mentioned in section 2.1 the treatments applied to the MODIS observations before AOD data assimilation are different among the members.

Verification with AOD products from other sensors was not included in this study because available products either have much smaller daily global coverage, (for example, the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and the Multi-angle Imaging SpectroRadiometer (MISR)), or are insufficiently characterized (e.g., Polar Multi-sensor Aerosol Product (PMAp)). The verification using the DA-quality MODIS product provides complementary spatial context to the AERONET comparison which is limited by the selective placement of the AERONET sites (Shi et al. 2011b).

Root mean square error (RMSE) incorporates both bias and variance information, and was used as a major metric for model validation in the first ICAP-MME paper (Sessions, et al., 2015). We continue to use this metric in this updated study, but with more recent model data and with two

more dust members. Other core verification metrics include mean error, mean absolute error, and coefficient of determination (r^2). Definitions of terms used in this paper are provided in Appendix B.

Since the ICAP-MME is run daily at 00Z for forecast out to 5 days, validations of the so-called "6-hr" or "72-hr forecasts" in this paper would be based on the forecast runs initialized at 00Z. This notation is also used in forecast error estimates for forecasts with different forecast lengths. Given AERONET and MODIS data are only available during local day-time, this corresponds to 6-24 hr of forecast time for any data day moving from the American continents, Atlantic, Europe and Africa, to Asia and Pacific in sequence. This gives American continents a beneficial regional verification bias, but we do not think this will impact any of our key results. This limitation is the same as in Sessions et al. (2015). Also for historical technical reasons, ECMWF did not report an analysis field of AOD at 00Z prior to January 2017. Thus, the 6-hr forecast valid at 00Z from all models with AOD data assimilation is used to approximate their analysis AODs.

3. ICAP-MME performance for June 2016-May 2017

3.1 Verification with AERONET AOD

Tables 2 and 3 provide total AOD and dust RMSE of all models (individual models and MME) from their 6-hr and 72-hr forecasts respectively at each AERONET site. Fig. 2 presents these RMSEs and additionally RMSEs for the fine and coarse AODs against each site's mean AOD. RMSE values for the fine and coarse AODs can be found in the Table S1 and S2 respectively. Similar to the earlier evaluation findings (Sessions et al., 2015) for total and coarse AODs, the ICAP-MME RMSE is either the leader or the second best in RMSE in nearly all cases. For fine-mode AOD, the ICAP-MME sometimes ranks third, however, the RMSE difference is less than

0.02 for both the 6-hr and 72-hr forecasts compared to the top ranked models, except for the Monterey site.

The MME dust forecast based on 7 dust members is not as skillful regarding ranking as in the previous evaluation, where the 5 member dust ensemble ranked the 1st for almost all dust sites and forecast hours in RMSE. But MME still ranks the 1st and 2nd for 6 out of 10 sites, and its RMSE is very close to the top ranked models for sites where it ranks the 3rd or 4th place over the other 4 sites (RMSE difference less than 0.02 for the 6-hr forecast and less than 0.04 for the 72-hr forecast).

Based on the slope of the RMSE versus AOD value linear regression for each site in Fig. 2, the RMSEs of ICAP-MME 6-hr forecast are approximately 50% of the yearly mean AOD value. Dust AOD forecasting is better than the individual fine and coarse mode AOD components, with its RMSE about one-third of the mean AOD. The RMSEs of the 72-hr forecast are about 10% larger for the AODs in each size mode compared to the 6-hr forecast. These results are similar to the previous findings for total, fine and coarse AODs (Sessions et al. 2015).

Overall, the models have reasonable correlation and consistency across the AERONET sites. Capo Verde, a very widely used benchmark site for African dust, consistently has RMSE approximately one-third of its annual mean for total and coarse-mode AOD, below the average of 50% for all sites for the 6-hr forecasts. Sea salt aerosol particles can be a contributor to coarse-mode AOD at this site, but dust is the dominant coarse-mode species. To allow for all ICAP models to be verified, model dust AOD instead of coarse-mode AOD (only available from 4 models) is verified again AERONET coarse-mode AOD. There is generally good agreement on dust and total AOD time series between observations and models (Fig. 3). Overall, the ICAP-

MME has a relatively better combined RMSE, correlation and dynamic range of data (95, 90, 75, 50, 25, 10 and 5th percentiles of data) in dust, fine-mode and total AOD in both the 6-hr and 72-hr forecasts compared to the individual models. Most background sites performed equally well, except Monterey on the central coast of California, with RMSE values approximately twice the mean AOD. Monterey, in normal years, is quite clean and has some of the best air quality in the United States. However, the local Soberanes wild fire that occurred in July-October 2016 makes 2016 an unusual year. The biomass burning smoke inventories used by models may not provide correct smoke fluxes, with large errors in both amplitude and pattern (e.g. Goodrick et al. 2013). Also the site is influenced by the sea-breeze and other meso-scale systems, which may not be well represented in the global models. As a result, the high smoke aerosol level and its large spatial and temporal variability was a big challenge for all global aerosol models. As an outlier, this site is excluded in the linear regression of RMSE against AOD in Fig. 2.

Different from the last evaluation, the remote oceanic site Crozet Island located in the "Roaring Forties" (high wind area) of the Southern Ocean, is now replaced with Amsterdam Island, which is just off the strong climatological wind belt in the Southern Indian Ocean. The performance of the ICAP models over Amsterdam Island is similar to other background sites with no indication of the significant overestimation of sea salt production that was found for Crozet Island in Sessions et al. (2015). However, the ICAP MME mean sea salt AOD values over the Southern Ocean between the recent years and earlier years (including 2012 when Sessions (2015)'s evaluation was based) are comparable from the daily AOD distribution maps shown online. These indicate that the ICAP models have problems specific to the high-wind, high sea-salt production areas of the Southern Ocean, and suggest a requirement for better sea salt

parameterizations and potential problems with the widely used exponential relationship between surface wind and sea salt production in current aerosol models.

Also of note is the Beijing site for which the models continue to demonstrate poor forecasting skill, with RMSE values for total AOD similar to the mean AOD. This is owing to the strong inversions and complex secondary production processes that result in thick haze that frequent the area (e.g., Guo et al., 2014; Zhang et al., 2014). However, the RMSE values at this site are smaller than during the 2012 time period used in the first ICAP-MME paper (also see section 4). The relatively poorer skill at fine-mode AOD compared to dust/coarse mode is the main contributor to the overall large RMSE (Fig. 4). Fine mode AOD from AERONET exhibits great temporal variability on day-to-week time scales. All the individual core models and the MME have difficulty in capturing this large variability in fine-mode AOD. They tend to overestimate in clean conditions and underestimate in highly polluted conditions. It is very common for global aerosol models to yield a smaller range of AOD values compared to observations and fail to capture the magnitude of big events (Kinne et al., 2006; Sessions et al., 2015). This may be due to emissions or aerosol processes that are not fully understood or characterized. This behavior is also expected mathematically based on the spatial scales of models and observations, but is enhanced by other properties of models, for example, representations of surface gustiness, orographic flows and other boundary layer processes relevant to aerosol sources and sinks. This behavior can also be seen in global aerosol reanalysis products (Lynch et al., 2016; Randles et al., 2017; Yumimoto et al., 2017).

All of the four core multi-species models have AOD data assimilation. However, even with DA models did not appear to reproduce the high AODs above the 90th percentiles for Beijing. This is because a) satellite retrievals are also challenged by the complicated aerosol and land

environment over East Asia often flagging thick haze events as cloud, and b) variational data assimilation can have difficulty spreading what little observational data is available, and specifically reproducing strong gradients found near surface sources (Rubin et al., 2017). There can be coexistence of dust and pollution particles of different sizes, sometimes the AOD is too high for valid retrievals (Shi et al., 2015), and sometimes interaction and transport of aerosols with cloud and/or fog prevents retrievals (Eck et al., 2018). A severe haze event that occurred on October 13, 2016 is an example for which all the ICAP models failed to predict the high AOD. As shown in the Terra MODIS true color image (Fig. 4i), heavy haze is covering northeast China where Beijing is located. The haze was so thick that no valid AOD retrievals were available (Fig. 4j), possibly due to a combination of the low cloud masks, the upper limit of AOD retrieval at 5 and the inland water mask in the regular MODIS retrieval algorithm. A modified MODIS retrieval algorithm targeting high AOD situations, is being developed and tested, shedding light for improvement of satellite AOD product for cases like this (Shi et al., 2018). However it may take time for the research algorithm to mature and be incorporated into the operational retrieval algorithms for modeling purposes. Regardless, with current available retrieval products, the nearest quality observations no doubt have lower AODs than the peak regions. Since total AOD is the common variable being assimilated, there are no constraints on fine/coarse aerosol partitioning or aerosol speciation. Even if the total AOD product is perfect in quality, the resulting speciated AODs and modal AODs can be very different.

The bias of coarse-mode AOD or dust AOD is much smaller compared to the bias of fine-mode AOD, and the range of dust AOD in MME is comparable to that of the observations. However, correlations for dust AOD are low, suggesting the models have difficulty producing the timing of dust events. Those factors combined result in poor verification scores for ICAP-MME over

Beijing, although it still ranks the top among all models with respect to RMSE, correlations and dynamic range of AODs. Improvements in data assimilation systems currently under development (e.g., Rubin et al., 2017) show significant promise for improving aerosol prediction in these conditions.

3.2 Verification with DA-quality MODIS AOD

In order to globally evaluate the performance of the ICAP models, total AOD at 550 nm from the individual ICAP models and the MME are compared with the DA-quality MODIS C6 product. The geographic distribution of MODIS DA-quality AOD averaged over the one year study period is presented in Fig. 5, as well as the pairwise total AOD from the ICAP-MME. The global distribution of the total number of 6-hourly 1°x1° MODIS observations is also shown. The DA-quality product includes albedo filtering based on MODIS 16-day surface albedo/Bidirectional Reflectance distribution function (BRDF) product (MCD43C3; Schaaf et al., 2002), excluding areas with low signal-to-noise as diagnosed using a 10+ year dataset comparing AERONET and MODIS (Hyer et al., 2011). Coverage over bright areas is improved by using MODIS Deep Blue retrievals, but many bright surfaces are still excluded in the DA-quality product. Areas with high cloud coverage, including the ITCZ, the Maritime Continent and the subtropical stratus cloud deck regions, have relatively less data. Cloudy conditions, problems retrieving over snow, and polar night limit retrievals of AOD at high northern latitudes. Over the Southern Ocean, MODIS AOD retrievals exhibit an anomaly which has been shown to be partially but not entirely attributable to undetected cloud (Toth et al., 2013; Christensen et al., 2015); because of this, retrievals south of 40°S are excluded from this analysis.

The global distribution of mean total AOD from the ICAP-MME looks similar to that of the DA-quality MODIS AOD. Prominent high AOD features exist over dust-influenced regions, including north Africa, Sahel, Arabian Peninsula and central Asia; biomass-burning-dominated central and South Africa, South America, Peninsular Southeast Asia and Siberia; and East Asia and India, which are impacted year round by pollution and seasonally by dust and biomass burning. There are also areas of significant disagreement between the MODIS C6 DA-quality dataset and the ICAP-MME. For example, ICAP-MME total AOD is lower over East Asia, India and Siberia, and higher over central Asia, Arabian Peninsula, indicating biases relative to MODIS C6 (also Fig. 7). ICAP-MME is also relatively high over the western U.S., which may reflect differences between MODIS Collection 5 AOD assimilated into the ICAP models and Collection 6 MODIS AOD used for this comparison (see e.g. Levy et al. 2013; Sayer et al., 2014). The Collection 6 MODIS Deep Blue products also have a documented problem with elevated terrain that can be seen as a low bias in MODIS AOD over Iran and other elevated areas in Asia and North America. This problem is corrected in the newer Collection 6.1 version (https://modis-atmosphere.gsfc.nasa.gov/sites/default/files/ModAtmo/modis_deep_blue_c61_changes2.pdf), but the newer data were not available in time to use in this study.

Fig. 6 shows global distributions of interquartile values (median, 25th and 75th percentiles) of the MODIS DA-quality AOD for the one year study period, and ratios of the same quantities from the 6-hr and 72-hr forecasts of the ICAP-MME to the MODIS AOD. "Analysis mode" and "forecast mode" refer to the 6-hr forecasts and 72-hr forecasts from now on. The median of MODIS AOD is very similar to the mean (Fig. 5b) except over Siberia, where the mean is much higher than the median, likely because the sample size is relatively small (less than 60) and the

mean is dominated by some high AOD observations associated with large biomass burning events. Similarly, the ICAP-MME median looks very much like the mean except over Siberia (not shown). Consistent with the aforementioned bias analysis, the ICAP-MME median AOD is higher than MODIS over central Asia and the western U.S., and lower over East Asia, India and Siberia. There is a very clear tendency for the ICAP-MME to be lower than MODIS in the 75th percentile AOD over the globe, except for the high-biased regions. The higher AOD in these regions could be a result of differences between the satellite data assimilated into the model and the verification dataset, as discussed above. In the 25th percentile AOD, the ICAP-MME is generally higher than MODIS except in low-biased regions. These results are approximately true for both the analysis and the forecast modes, except that the biased regions tend to be slightly more biased in the forecast mode. Similar patterns are seen for all of the individual models (Fig. S3). This means that, in general, the ICAP-MME and all the contributing global models tend to overestimate in clean conditions and underestimate in severe aerosol conditions. This result is consistent with the validation with AERONET in Section 3.1 and other global aerosol modeling studies (e.g., Kinne et al., 2006; Sessions et al., 2015; Lynch et al., 2016). Fig. 7 shows global distributions of biases, RMSEs, and the coefficients of determination of the four core models and ICAP-MME for their analysis mode (from 6-hr forecast). The validation patterns for the forecast mode (72-hr forecast) look similar; except that biases and RMSEs are slightly larger and the correlations are slightly lower (Fig. S2). There are consistent low biases across the models over Siberia and India, especially the southern foothill of the Himalayas, most likely resulting from under-prediction of smoke over Siberia and anthropogenic and biogenic aerosols over India in the models. Consistent high biases are found over central and East Asia dust-dominant regions. Other regions tend to have mixed results. It is also noted that Model 1 has a slight high bias over

the entire globe, much of which disappears over water in the forecast mode (Fig. S2). This implies that the AOD observations assimilated into Model 1 were slightly higher than the DA-quality MODIS C6 product used here.

RMSEs are commonly higher over the biased regions, which are often the climatologically high AOD regions. Correlations are high over the oceanic areas where large-scale transport of dust, smoke and pollutions occur downwind of its continental sources. Over land, r^2 is relatively lower overall because land is the main source of aerosols studied here except sea salt, and there are much larger uncertainties and stronger gradients due to local aerosol sources than large-scale transport events in aerosol modeling. AOD data assimilation helps improve r^2 over land, but it helps more over ocean, because of longer transport times as well as the higher signal/noise ratio of AOD retrieval over ocean (e.g., Levy, et al., 2005). High overland r^2 occurs over the pure (not mixed with other aerosol species) biomass-burning dominant regions, including South Africa, South America, Southeast Asia and the boreal burning regions, resulting from the fact that all ICAP models (see Table 1) use smoke emission inventories based on satellite observations which are updated in near real time. Other sources, including dust and sea salt emissions, are generally parameterized based on limited field measurements. Emissions for anthropogenic and biogenic sources have even more degrees of freedom for uncertainty given their complex chemistry and interactions with meteorology. Thus correlations are low in these source regions. Because of small dynamic ranges of AOD in the most remote regions, correlations are also low in areas far from aerosol sources or transport paths, e.g., subtropical Pacific and Indian oceans,.

Consistent with the validation result against AERONET, the ICAP-MME performs the best among all the ICAP full-species models verified with the DA-quality MODIS total AOD. The ICAP-MME global mean absolute error and RMSE are the smallest, with similar magnitude to

these of the individual models. Correlation of ICAP-MME is significantly higher than individual models, with global mean r^2 higher than individual models for both the analysis and forecast modes. The r^2 increases from 0.42 on average for individual models to 0.53 for the MME in the analysis mode. In the forecast mode, the single-model r^2 averages 0.26 versus 0.35 for the MME. This can be related to situations in which the MME captures events missed in some models but captured by other models. As expected, the performance of all models is worse in their forecast modes (Fig. S2) as they move away from the time of data assimilation. Overall, with some exceptions, the ICAP core models have similar performance regionally and globally, with small divergence among the models for both the analysis and the forecast modes, despite great diversity among systems. The consistently challenging regions across the ICAP models are dusty regions over land and regions with two or more dominant species. These include India and East Asia, influenced by pollution and dust, and the Sahel, influenced by dust and biomass burning smoke.

4. Ensemble mean and spread of ICAP models and potential for probabilistic prediction

One of the goals of ICAP is to advance probabilistic aerosol forecasting, which provides aerosol forecasts with associated uncertainties. This is an advantage over deterministic forecasts, especially for severe events, in that the predictions have an associated confidence level. It is analogous to NWP and TC ensemble predictions, where severe precipitation or temperature events or TCs are predicted with certain possibility levels of hit or miss for a location. If predictions from all individual models converge, this indicates a prediction with high confidence or high possibility. This probabilistic prediction facilitates better decisions in preparation for such severe weather events. Similarly, probabilistic predictions for severe aerosol events, e.g., pollution, dust and biomass burning smoke events, are desired, motivating both single and multi-

model aerosol ensembles. The utility of the ensemble systems for such a purpose will be evaluated. AOD verification against observations in Section 3 shows the ICAP-MME consensus and diversity of model performance in analysis and forecast modes. In order to make use of the information for probabilistic prediction, it is necessary to quantify the mean and spread of the models and evaluate the usefulness of these variables for forecast uncertainty estimates. As defined above, ensemble mean is the unweighted arithmetic mean of ensemble members, and ensemble spread is estimated using the standard deviation of ensemble members.

4.1 Ensemble mean and spread of the ICAP models in analysis and forecast modes

Fig. 8 presents the global distributions of yearly average ICAP ensemble mean and ensemble spread of total, fine and coarse-mode AODs for the analysis mode and the differences between the forecast and analysis modes. As expected, ensemble spread tends to be large over high AOD regions and small over low AOD regions. This is true for both the fine and coarse modes and the total AOD. However, different behavior is observed in India, where the mean fine and total AODs are comparable to those over East Asia and southern Africa, but the ensemble spreads are much smaller. This could be a result of consistent low bias (as shown in Fig. 6 and 7) and less variability over India across the ICAP models. The impact on the capability of ICAP-MME for regional probabilistic predictions will be discussed in section 4.2. Compared to the analysis mode, the total AOD ensemble mean for the forecast mode is smaller overall, mainly attributed to smaller fine-mode AOD (about 10% decrease). The observed ensemble mean decrease in the forecast mode occurs over biomass-burning impacted regions including South America, Sahel, South Africa, the Maritime Continent, Siberia, and the heavily polluted North India, suggesting insufficient emissions in the forecast mode. There are slight increases over North Africa, Australia and Arabian Peninsula, and slight decrease over subtropical north Atlantic in coarse-

mode AOD in the forecast mode, suggesting possible overestimation of dust emission overall and excessive removal over water.

The ensemble spread is generally larger for the forecast modes compared to the analysis modes, in which AOD is constrained by data assimilation in varying degrees in these models. Some deviations include the western United States, the Andes of South America and the Maritime continent for total and fine-mode AODs, where ensemble spread is smaller in the forecast mode than the analysis mode. This could be attributed to the diversities of the AOD data and/or the pre-treatment of these data that were assimilated into these models in the analysis mode. After all, large differences are found in satellite AOD products over these mountainous regions because of different capabilities of dealing with high-reflective and varying surface conditions in retrieval algorithms (Shi et al., 2011; Loria-Salazar et al., 2016). An additional contributor could be divergence in model meteorological variables that impact aerosol processes. For example, precipitation, controlling aerosol wet removal, can be more divergent in the analysis mode than the forecast mode in the NWP models, given the differences between satellite precipitation products assimilated and NWP models (Ebert et al., 2007). By using satellite derived versus model precipitation in an aerosol modeling study, Xian et al. (2009) found significant difference in AOD levels over many regions, including Andes and Maritime continent. The ensemble spread of fine-mode AOD is also smaller in the forecast mode than the analysis mode over remote oceanic areas, which is associated with slightly smaller ensemble mean total/fine AOD. This is because one of the four data assimilation models has slightly higher background AOD over ocean in its analysis mode, but similar background AOD in the forecast mode when compared to the other models. This is consistent with the comparison to the MODIS DA-quality product shown in Section. 3.2 (Fig. 7 and S3).

Fig. 9 shows global distributions of yearly average ICAP ensemble mean and ensemble spread of dust AOD from the DA models at their analysis mode and the differences from their forecast mode, and the differences from all models at the analysis and forecast modes. Error growth from analysis to forecast mode is a function of NWP forecast errors, errors in prediction of sources and sinks, and relaxation from aerosol analysis state (for models with aerosol data assimilation). Models without aerosol data assimilation are evaluated here together with the DA models at 6-hr forecasts for probabilistic forecast purposes. The dust AOD ensemble mean based on the 5 DA models at the analysis mode (Fig. 9a), shows the main dust active regions including North Africa, Arabian Peninsula, central Asia, south Asia, Australia, western U.S.A., and southwest South America and their downwind regions. As expected, high dust AOD regions also exhibit high ensemble spread. The general tendency for all models at both their analysis and forecast modes and the DA models at their forecast mode to have higher dust AOD over North Africa and Arabian Peninsula, and lower dust AOD over South Asia, suggest possible excessive emission over North Africa and Arabian Peninsula, and insufficient emission over South Asia. There is also a tendency of lower dust AOD in the forecast modes over the subtropical Atlantic, which is the long-range transport region of African dust, indicating excessive removal of dust over water in the models.

Since AOD is constrained with satellite retrieved AOD at the analysis time in DA models, ensemble spread of the DA models in the analysis mode is reduced overall compared to all models and their forecast modes (Fig. 9d,f,h). The spread of the DA models is discernably reduced more over water than over land in the analysis mode. This is because satellite AOD products have much larger uncertainties over land than over water (e.g., Levy et al., 2013; Zhang et al., 2008; Hyer et al., 2011). Moreover, there is much less DA-quality AOD data over bright

desert for models to assimilate (e.g., Fig. 5c). Although some models assimilate satellite AOD products with coverage over desert, others using different AOD products or with very strict QA/QC processes may not have much data to assimilate over some desert areas. Also notable is the small difference in ensemble spread over East and South Asia dust areas for the DA and all models, and for analysis and forecast modes. This reflects the challenge of dust modeling and AOD retrieval in this complicated aerosol environment, in which dust and various kinds of pollutions are mixed, and complex chemistry of precursors and secondary organic aerosols convolve with meteorology (e.g., Zhang et al., 2014).

4.2 Estimation of ICAP-MME absolute forecast error

In order to evaluate the usefulness of ICAP-MME for probabilistic forecasts, we first explore the relationships between the possible predictors and the ICAP-MME absolute AOD forecast errors. The predictors examined include ensemble mean, ensemble spread and forecast AOD change (defined as the forecasted change of AOD in 24 hours). Linear correlations between ensemble mean and error, ensemble spread and error, and forecast AOD change and error are calculated for modal AODs and dust AOD respectively. Some statistically significant correlations for the dominant aerosol modes are found over most of the selected 21 AERONET sites for the 6-hr and the 72-hr forecasts (coefficients of determination for the 72-hr forecasts are shown in Table S2, and those for the 6-hr forecasts are similar). In general, correlations between ensemble mean and forecast error are slightly higher than correlations between ensemble spread and forecast error. However, weak or insignificant correlations are found between forecast AOD change and absolute forecast error for the 6-hr and 72-hr forecasts (not shown).

For dust AOD, ensemble mean and spread show statistically significant correlations with consensus forecast error over most of the dust sites for both the 6-hr and the 72-hr forecasts (Table 4). No correlation or low correlation is found over Gandhi College, Kanpur and Yonsei University sites, indicating ensemble mean and spread have little or limited skill in ensemble mean error estimates for these sites. However, it is also noted that for Gandhi College, most available AERONET observations coincide with periods of calm winds and minimal dust production, and strong dust events reflected in the ICAP-MME cannot be verified with AERONET (not shown). Similarly, there is no correlation between ensemble mean/spread and forecast error over Gandhi College and Kanpur for total and fine-mode AODs (Table S3), mostly due to the consistent low biases and small ensemble spread among the models.

It is also found that, for all sites, coefficient of determinations (r^2) between the ensemble mean and spread are high, on the order of $r^2=0.4$ -0.9 for total, fine and coarse AOD forecasts (Table S1) and on the similar order for dust AOD forecasts. This is consistent with Fig. 8 and 9.

Our analysis shows that forecast error is correlated slightly more with ensemble mean than ensemble spread in the ICAP MME aerosol forecasts. This indicates large room for improvement in global aerosol modelling, and before these models reach maturity other factors may also play a role in ensemble error estimates. We expect that with continuous development of individual models, the ensemble spread will carry more weight in the error forecast model, as is seen in the evolution of error forecast model for tropical cyclone (TC) track and intensity ensemble forecasts for the past decade.

Following the studies on prediction of consensus TC track and intensity forecast errors (Georss, 2006; Georss and Sampson, 2013), we regress absolute AOD forecast error on the related

predictors, including ensemble mean and ensemble spread, and derive regression models for ICAP consensus forecast error based on all available model and AERONET data from the 21 representative sites for total AOD (Table 5) and the 10 dusty sites for dust AOD (Table 6). For the total AOD forecast, ensemble mean is found to be the leading predictor, with r^2 ranging from 0.27 to 0.22 monotonically corresponding to forecast lengths of 6-hr, 24-hr, 48-hr, 76-hr and 96-hr. Ensemble spread is the second predictor, with r^2 varying between 0.16 and 0.22 for forecasts with the same forecast lengths. The forecast error prediction based on multivariate linear regressions of both ensemble mean and spread yields the best correlation with forecast error. The addition of ensemble spread adds only a small skill improvement on top of the ensemble mean to the forecast error estimate though.

Similar regression result is found for dust AOD forecast error, in which ensemble mean is the leading predictor. The forecast error prediction based on multivariate regressions of both the ensemble mean and spread again yields the best correlations for 6-hr to 96-hr forecasts although linear regression based on solely ensemble mean yield the same correlations for some forecast lengths. Comparisons of predicted total/dust AOD errors using the multivariate regression models and the ICAP-MME forecast error for the 72-hr forecasts are shown in Fig. 10 as an example. Time series of error estimates of dust AOD at a relatively skillful site, Capo Verde, and total AOD at a less skillful site, Beijing, are given in Fig. 11. There are times that absolute errors are much larger than those predicted. These points correspond to the cases that all models are consistently low biased and with small spreads. These common issues warrant model improvements, but the results here demonstrate the potentials of such error regression models for applications in probabilistic AOD forecasts.

The correlation relation between AOD absolute forecast error and ensemble mean/spread shows strong regional differences, which might be relevant to model skill in resolving different dominant species (e.g., dust, smoke and pollution haze) and/or regional meteorology that impacts aerosol processes. Similar error forecast models can also be developed for individual regionally-representative sites or regions and for different size-modes (fine vs coarse) for potential applications in the future. Each individual model's contribution to the ICAP-MME is evaluated by removing one model from the MME and quantifying changes in the one-year mean absolute forecast error. The results are mixed for different models and different sites for dust AOD forecasts (Table 7). The percentage changes for all sites are averaged in order to give an overall evaluation. For dust AOD forecasts, the mean absolute forecast errors (AFE) increase about 5% with removal of Model 5 and 7 (slight more error increase), and about 2-3% with removal of Model 2 for the 6-hr and 72-hr forecasts, suggesting that these three models are contributing positively to the consensus mean. However the mean AFE decreases 6%/2% for the 6-hr/72-hr forecast, with removal of Model 6, suggesting the model is contributing slightly negatively to the consensus mean. The other three models have mixed result: AFE slightly increases for one (analysis or forecast) mode and slightly decreases for the other mode, or with AFE unchanged, indicating their contributions to the MME are approximately neutral. Similar evaluations are done for the total, fine-mode and coarse-mode AOD forecasts with removal of one of the four full-species models. It is found that all the four core models contribute positively to the ICAP-MME total AOD forecasts, with a 2%-3% average (of all 21 AERONET sites) reduction in RMSE from removing each of the four models in the 6-hr and 72-hr forecasts (Table S4). However, for the MME fine-mode AOD forecasts, Model 1 contributes negatively most likely due to its slightly high bias over relatively clean regions (see also Fig. 7), while the other three

models contribute positively (Table S5). For the MME coarse-mode AOD forecasts, contributions are positive from Model 2 and Model 3, neutral from Model 1 and slightly negative from Model 4 (Table S6). These results suggest that the full-species models perform similarly in terms of total AOD in general, but their performances in terms of fine- and coarse-mode AODs are different. This also reflects the fact that data assimilation of total AOD can help constrain total AOD, but it does not constrain contributions from different aerosol species. We acknowledge that this evaluation of each individual model's contribution to the MME is quite arbitrary, yet it reflects the complex impact on the MME performance of adding more independent ensemble members. With rapid evolution in the individual member models these numbers are expected to change.

5. Evolution of ICAP-MME performance over 2012-2017

Since its initial operation in 2011, ICAP-MME has incorporated a few more deterministic global aerosol models (Fig. 1), and numerous updates have been implemented on individual models by the contributing centers. Those updates include adding new species, e.g., organic aerosols and nitrate aerosols, and expanding from a single dust species to a multi-bin dust representation, as well as updates of aerosol processes, e.g., inventories for emissions and parameterizations for removals. Regarding aerosol data assimilation, updates include new and improved AOD products for DA and/or changes in their treatments prior to DA, or even major changes from no DA to DA. Furthermore, the underlying NWP models from all of these centers have also seen updates, ranging from finer spatial and temporal resolution, better physics and dynamics, to additional observational data and advanced methodologies for DA. Whether these updates improved the ICAP-MME performance over the years is examined here.

Fig. 12 illustrates the evolution of ICAP-MME performance in terms of 550 nm total AOD RMSE at all the selected AERONET sites for the 6-hr forecasts. The evolution of ICAP-MME performance in terms of fine-mode and coarse-mode AOD RMSEs are provided in Figures S3 and S4, respectively. Interannual variability in the performance of MME is noted for many sites, Singapore and Monterey being the most obvious two with extremely large variability within the 2012 to 2017 study period. Consistent with the result of Sessions et al., (2015) and Fig. 2, the RMSE generally increases with AOD. The anomalously high RMSEs in 2015 and 2016 for the two sites, respectively, are associated with high fine-mode AODs and high variability resulting from severe wild fire conditions. Singapore was impacted by the particularly strong and wide-spread biomass burning events across the Maritime Continent in 2015 due to a strong El Nino (Fanin and van der Werf, 2017; Huijnen et al., 2016; Tacconi, 2016). Whereas Monterey, California, which is typically pristinely clean, was influenced from time to time by smoke from wild fires lasting over two months in the nearby area in its dry season in 2016. As mentioned earlier and in other studies (Kinne et al., 2006), global models tend to underestimate extremely high AOD events, which leads to anomalously high RMSE in unusual years. A similar performance pattern is found for 72-hr forecasts, except for slightly higher RMSEs (not shown).

A significant decreasing trend in RMSE is present for Beijing, where ICAP-MME RMSE is reduced by half from 2012 to 2017 (from 0.64 to 0.30). This RMSE decrease is associated with decreases in the yearly means of total AOD observed by AERONET over Beijing for the study period. The decrease in total AOD is consistent with reported negative trends found in other studies using satellite AOD retrievals over East Asia (Zhang et al., 2017). The main contributor to the total AOD decrease is the decrease in fine-mode AOD with similar magnitude, with only a very slight decrease in dust levels over the years (Fig. 13 and Fig. S3, S4). Time evolution of the

fine-mode AOD RMSEs from the four core models and the ICAP-MME show that all the ICAP models have decreasing trends of varying magnitude in fine-mode AOD RMSE over Beijing (Fig. 13).

There are no significant changes in mean dust levels and variance over Beijing. However, performance of all the individual models in terms of dust forecast is more divergent among the models and over the years. ICAP-MME dust AOD RMSE is not always the lowest, but is relatively stable compared to individual models over the years. This is one of the benefits of multi-model ensembles. A similar conclusion can also be made with respect to regional performances. Individual models perform differently regionally. One model may perform better in some regions, but worse in other regions compared to other models. But ICAP-MME has more stable performance across all regions.

There is a tendency for smaller RMSE, especially in fine-mode AOD, over other Asian sites as well. Small decreasing trends in RMSE of total and fine-mode AOD are discernable for Kanpur and Gandhi College, the two Indian sites (Fig. 12 and Fig. S4). Different from Beijing, there is no clear trend in the yearly mean total and fine AODs and their standard deviations, suggesting the decreasing trend is a result of model improvements, especially in fine-mode AOD forecasts, over the region from 2012 to 2017 (Fig. 13). Yonsei University, Korea, and Chiang Mai Met Station, Thai, also show decreased fine-mode AOD RMSEs without significant decreases in their annual mean fine-mode AODs (Fig. S4), indicating model improvements in fine-mode AOD forecasts over the years. It is known that some models incorporated organic aerosols and/or nitrate aerosols in the fine-mode, and updated emission inventories, which would improve the low bias over India, East and Southeast Asia, where severe anthropogenic pollutions often occur. Additionally, one of the four core models (other three had DA since ICAP inception)

incorporated AOD data assimilation in the middle of 2016, which may have also contributed to the RMSE improvement in the recent two years.

No significant trends in ICAP-MME performance in terms of total AOD RMSE are found for other sites. Biomass burning and dust impacted sites tend to have large interannual variabilities in terms of AOD RMSE, mean and standard deviation because of the nature of these events. This may have blocked weak signals of model improvement if there are any. It is difficult to detect RMSE trends at background sites due to a small average and range of AOD.

Finally, the rankings of ICAP-MME among all the models in terms of total AOD RMSE of the 6-hr and the 72-hr forecasts for all the sites over 2012-2017 is shown in Fig. 14. As expected, ICAP-MME is either the 1st or 2nd place for most sites and years for both the analysis and forecast modes, indicating MME performance is good and stable over the years. Individual models could rank 1st for some sites/regions and years, but none of the individual models have high and stable rankings like the MME over time (Fig. S6). This is understandable as global operational aerosol models evolve quickly and the dynamic nature of significant aerosol events, such as related to large wildfire outbreaks or heavy dust seasons. When there is a model upgrade, there is usually abrupt performance change associated with it. An upgrade can impact some regions more than other regions or some aerosol species more than other species. Sometimes it may not be model upgrade, but just model physics that can result in a good simulation for one scenario but bad simulation on another. In the long run, the MME wins due to its averaging nature. Similar behavior of multi-model ensembles is also observed in the Tropical cyclone track and intensity forecasts where consensus prediction wins over individual models over a longer time span (e.g., DeMaria et al., 2014). This is also why a consensus mean, i.e., even-weighting for all the participating models, is adopted in the ICAP-MME.

6. Discussions and conclusions

This paper provides an update on the International Cooperative for Aerosol Prediction (ICAP) global operational aerosol multi-model ensemble (MME) AOD consensus product. Compared to the first ICAP-MME analysis (Sessions et al., 2015), the multi-species models are still the four models: ECMWF CAMS, JMA MASINGAR, NASA GEOS-5, NRL NAAPS, while the dust models have expanded from the original five (aforementioned four, plus NOAA NGAC) to include additional BSC MONARCH and UKMO unified dust model, making seven dust models in total in this study. The newer ICAP members, namely NOAA full-species NGAC, Meteo France MOCAGE and FMI SILAM, are not included in this study because of shorter data records. A recent full year of data, from 1 June 2016 to 31 May 2017, is used for detailed ICAP-MME performance statistics compared to observations and to evaluate the usefulness of ICAP-MME for probabilistic forecasts. The evolution of the ICAP-MME performance during 2012-2017 is also examined. We expect rapid evolution in the individual member models based on the results shown here and similar exercises with ICAP-MME products. So the error metrics may be out of date for the better by the time this article is published. The current state of the ICAP-MME, and the similarities and differences between these findings and the initial ICAP-MME evaluation made with the first year of ICAP data, which was five years older (Sessions et al., 2015), are documented by our results, along with the usefulness of the ICAP-MME for aerosol probabilistic forecast. The main conclusions from this analysis are listed here as follows:

1. ICAP-MME ranks 1st overall among all individual models in terms of overall RMSE, coefficient of determination (r^2), and bias for both analysis and forecast modes for total, fine and coarse and dust AOD based on verifications against AERONET Version 3 L1.5

observational data and DA quality MODIS C6 product. This result is similar to the first ICAP-MME evaluation by Sessions et al. (2015).

2. In general, the AOD spread of models with data assimilation at their analysis mode is smaller than the AOD spread of all models at their analysis mode, which is smaller than the spread of all models at their forecast mode. This is true for total, fine, coarse and dust AODs over the globe, except over India and dusty East Asia region, where global models have common low biases and small spread for cases of heavy regional pollution and sometimes mixed dust. These regions remain a challenge for global models, although model skills at AOD forecasts for these regions have been improved in the past five years (see conclusion #4).

3. ICAP-MME ensemble mean and spread have skills for predicting absolute AOD forecast error globally, except for over India, where they have little correlation with forecast error. Multivariate regression models of absolute forecast error are derived based on both ensemble mean and spread for total and dust AOD forecasts at different forecast lengths. These regression models can potentially be applied for probabilistic AOD forecasts.

4. ICAP-MME performance in terms of modal AOD RMSEs of the investigated 21 regional representative sites over 2012-2017 shows a general tendency for model improvements in fine-mode AOD, especially over Asia. No significant improvement in coarse-mode AOD was found overall. Interannual variability in regions influenced by biomass burning smoke and dust may obscure small signals of potential model improvement.

5. ICAP-MME performance is stable and reliable over the years compared to individual models. AOD RMSE of the ICAP-MME is not always the lowest for a given species, site or year, but it is relatively low and stable. Individual models may perform better in some

regions/years/scenarios and worse in others and may experience abrupt performance changes associated with upgrades. Consensus MME wins in the long run because of its averaging nature of independent models.

Individual contributing centers have their own plans for future aerosol model developments, with the development focus depending on their customer needs and current model status. These plans may include addition of aerosol species, update of emission inventories, addition/update of aerosol data assimilation, increased model resolution, improved parameterization of physical, chemical and/or optical properties and processes. These future plans also stress requirements for aerosol observations in the context of the operational activities carried out at various centers (Benedetti et al., 2017).

Currently the ICAP-MME products and the evaluations of ICAP-MME performance are based on speciated or modal AODs because AOD has the most abundant observations and global coverage and it provides a big picture of column total amount of aerosols. The next big move for the ICAP MME is towards surface PM2.5 (Particulate Matter with aerodynamic diameter less than 2.5 µm) and PM10 (Particulate Matter with aerodynamic diameter less than 10 µm) ensembles. Data collected from global observational networks for these properties will be used for evaluations. The evaluations of these new ensemble properties would help the operational aerosol communities to identify issues relevant to surface/lower boundary layer properties and lead to potential improvements. In the future, aerosol vertical distributions can also be investigated through the ICAP-MME framework.

Data Availability:

The ICAP-MME modal and dust AOD product is available

http://usgodae.org/cgi-bin/datalist.pl?dset=nrl_icap_mme&summary=Go

The MODIS data-assimilation quality gridded AOD product, used for global-scale verification of ICAP-MME, is available in near real-time from NASA LANCE: http://dx.doi.org/10.5067/MODIS/MCDAODHD.NRT.006.

development has been supported by Joint Center for Satellite Data Assimilation, NOAA Modeling, Analysis, Prediction, and Projections program and National Weather Service. MONARCH development is supported by the Spanish Government. Development of the Met Office Unified Model is funded by a combination of the UK Public Weather Service and the Hadley Centre Climate Program supported by the Department of Business, Energy and Industrial Strategy (BEIS) and the Department for Environment, Food and Rural Affairs (Defra). MOCAGE development is supported by Centre National de Recherches Météorologiques (CNRM-GAME) of Météo-France and Centre National de la Recherche Scientifique (CNRS). Global SILAM version was developed under ASTREX, APTA, and GLORIA projects of Academy of Finland; the fire emission system was developed within the Academy IS4FIRES project.

Appendix A: Member model descriptions

A1. BSC MONARCH

The Multiscale Online Nonhydrostatic AtmospheRe CHemistry model (NMMB-MONARCH v1.0; Pérez et al., 2011; Haustein et al. 2012; Jorba et al., 2012; Spada et al., 2013; Badia et al., 2017), formerly known as NMMB/BSC-CTM, is a fully online integrated system for meso- to global-scale applications developed at the Barcelona Supercomputing Center (BSC). The model provides operational regional mineral dust forecasts for the World Meteorological Organization (WMO; https://dust.aemet.es/), and participates to the WMO Sand and Dust Storm Warning Advisory and Assessment System for Northern Africa-Middle East-Europe (http://sds-was.aemet.es/). Since 2012, the system contributes with global mineral dust and sea salt aerosol forecast to the multi model ensemble of ICAP at a resolution of 1.4° x 1° on 24 hybrid sigma-

pressure levels. NMMB-MONARCH v1.0 has been enhanced with a new hybrid sectional-bulk multicomponent mass-based aerosol module (Spada, 2015). The aerosol module is designed to provide short and medium range forecast of the atmospheric aerosols for a wide range of scales, with the option to adjust the complexity of the chemistry scheme as desired. The module describes the lifetime of dust, sea-salt, black carbon, organic matter (both primary and secondary), sulfate and nitrate aerosols. While a sectional approach is used for dust and sea-salt, a bulk description of the other aerosol species is adopted. The CB05 chemical mechanism (Yarwood, 2005) can be selected to solve the gas-phase chemistry or, alternatively, climatologies of the most important oxidants are used for simplified global aerosol runs and forecast. A simplified gas-aqueous-aerosol mechanism has been introduced in the module to account for the sulfur chemistry and a two-product scheme is used for the formation of secondary organic aerosols (Spada, 2015). An upgrade of the NMMB-MONARCH v1.0 ICAP aerosol forecast is planned for mid-2018. The system will provide forecast of mineral dust, sea salt, carbonaceous aerosols and sulfate at a resolution of 0.7º x 0.5º on 48 hybrid sigma-pressure levels. Global anthropogenic emissions from the AEROCOM-HTAP v2 dataset (Janssens-Maenhour et al., 2015) together with online MEGAN biogenic emissions (Guenther et al., 2016), and GFAS v1.2 biomass-burning analysis (Kaiser et al., 2012) will be used. Additionally, an aerosol data assimilation capability has been recently implemented in NMMB-MONARCH v1.0 (Di Tomaso et al., 2017). An ensemble-based data assimilation scheme (namely the local ensemble transform Kalman filter – LETKF) will be utilized in the near future to optimally combine model ensemble forecasts and observations, using a perturbed physics ensemble of NMMB-MONARCH v1.0. Results assimilating mineral dust optical depth derived from satellite retrievals (MODIS AOD Dark Target and Deep Blue) show a significant improvement of the forecast of mineral dust.

A2. Copernicus/ECMWF CAMS IFS

Starting in 2008, ECMWF has been providing daily aerosol forecasts including dust as part of the EU-funded projects GEMS, MACC and MACC-II and continuing operationally as part of the Copernicus Atmosphere Monitoring Service (CAMS), which provides predictions of global atmospheric composition and regional European air pollution. All data are publicly available online at http://www.copernicus-atmosphere.eu. The current model resolution is ~ 40 km with 60 vertical levels. A detailed description of the ECMWF forecast and analysis model including aerosol processes is given in Morcrette et al. (2009) and Benedetti et al. (2009). The initial package of ECMWF physical parameterizations dedicated to aerosol processes mainly follows the aerosol treatment in the LOA/LMD-Z (Laboratoire d'Optique Atmospherique/Laboratoire de Météorologie Dynamique) model (Boucher et al., 2002; Reddy et al., 2005). Five types of tropospheric aerosols are considered: sea salt, dust, organic and black carbon and sulfate aerosols. Prognostic aerosols of natural origin, such as mineral dust and sea salt are described using three size bins. For dust, bin limits are at 0.03, 0.55, 0.9 and 20 microns, while for sea salt bin limits are at 0.03, 0.5, 5 and 20 microns. Emissions of dust depend on the 10 m wind, soil moisture, the UV–visible component of the surface albedo and the fraction of land covered by vegetation when the surface is snow free. A correction to the 10 m wind to account for gustiness is also included (Morcrette et al., 2008). Sea salt emissions are diagnosed using a source function based on work by Guelle et al. (2001) and Schulz et al. (2004). In this formulation, wet sea salt mass fluxes at 80% relative humidity are integrated for the three size bins, merging work by Monahan et al. (1986) and Smith and Harrison (1998) between 2 and 4 mm. Sources for the other aerosol types, which are linked to emissions from domestic, industrial, power generation,

transport and shipping activities, are taken from the MACCity monthly mean climatology (Granier et al., 2011). Emissions of OM (organic matter), BC and SO2 linked to fire emissions are obtained using the Global Fire Assimilation System (GFAS) v1.2 based on MODIS satellite observations of fire radiative power, as described in Kaiser et al. (2012). In the absence of a chemical model of secondary organic aerosol production from anthropogenic VOCs, an additional source of OM proportional to anthropogenic CO emissions (as a proxy) is included. Several types of removal processes are considered: dry deposition including the turbulent transfer to the surface, gravitational settling, and wet deposition including rainout by large-scale and convective precipitation and washout of aerosol particles in and below the clouds. The wet and dry deposition schemes are standard, whereas the sedimentation of aerosols follows closely what was introduced by Tompkins (2005) for the sedimentation of ice particles. Hygroscopic effects are also considered for organic matter and black carbon aerosols.

MODIS AOD data at 550 nm are routinely assimilated in a 4-D Var framework which has been extended to include aerosol total mixing ratio as extra control variable (Benedetti et al., 2009). A variational bias correction for MODIS AOD is implemented based on the operational setup for assimilated radiances following the developments by Dee and Uppala (2009). The bias model for the MODIS data consists of a global constant that is adjusted variationally in the minimization based on the first-guess departures. Although simple, this bias correction works well in the sense that the MACC analysis matches well the de-biased MODIS observations. The observation error covariance matrix is assumed to be diagonal, to simplify the problem. The errors have been chosen based on the departure statistics and are prescribed as fixed values over land and ocean for the assimilated observations. The aerosol background error covariance matrix used for aerosol analysis was derived using the Parrish and Derber method

(also known as NMC (National Meteorological Center, now National Centers for Environmental Prediction) method; Parrish and Derber, 1992) as detailed by Benedetti and Fisher (2007). This method was long used for the definition of the background error statistics for the meteorological variables and is based on the assumption that the forecast differences between the 48 h and the 24 h forecasts are a good statistical proxy to estimate the model background errors. Since 2017, the METOP PMAp 550nm AOD product (https://navigator.eumetsat.int/product/EO:EUM:DAT:METOP:PMAP) has also been included in the assimilation in a similar way, except that errors provided with the product are used directly.

A3. JMA MASINGAR

The Model of Aerosol Species in the Global Atmosphere (MASINGAR) is an aerosol transport model developed at Meteorological Research Institute (MRI) of Japan Meteorological Agency (JMA) (Tanaka and Ogi, 2018; Tanaka et al., 2003). The aerosol model considers major tropospheric aerosol species including sulfate (and its precursors), black carbon (BC), organic aerosols (OA), sea salt, and mineral dust. Dust and sea salt aerosols are logarithmically divided into 10 discrete size bins from 0.1 to 10 μm in radius, while sulfate, BC and OA are assumed to have a lognormal size distribution and treated with total mixing ratio. The transport of aerosol is calculated with 3-dimensional semi-Lagrangian advection, subgrid vertical diffusion, convective transport, and gravitational settling. Removal processes of aerosol include rainout, washout, and dry deposition. The chemistry of sulfate production includes oxidation processes of dimethyl sulfide (DMS), sulfur dioxide (SO_2), sulfur trioxide (SO_3), and carbonyl sulfide (OCS) with oxidants (OH, H_2O_2, HO_2, and NO_3). The mixing ratios of the oxidants are taken from simulated monthly averaged fields from the output of MRI-CCM2 (Deushi and Shibata, 2011),

as described in Tanaka et al. (2003) and Tanaka and Ogi (2018). BC and OA include hydrophobic and hydrophilic components. It is assumed that the hydrophobic BC and OA gain hydrophilicity with an e-folding time of 1.2 days by aging processes, following Cooke et al. (1999). The secondary organic aerosol is assumed to be formed from 10% of monoterpene and 1.2% of isoprene emission calculated by MEGAN v2 used in the Chemistry-Climate Model Initiative (CCMI) project. (Sindelarova et al., 2014). Emission flux of sea salt aerosol is estimated by the formulation of Gong (2003) as a function of surface wind speed at 10 m altitude. Emission flux of dust is calculated as a function of the friction velocity, soil moisture, soil type, snow cover and vegetation cover described in Tanaka and Chiba (2005), which is based on the saltation-bombardment dust emission (Shao et al., 1996). Anthropogenic emissions of SO_2, BC, and OA (organic aerosols) are specified by the monthly MACCity emission inventory (Granier et al., 2011). Daily emissions of SO_2, BC, and OA from biomass burning are incorporated from the GFAS inventory obtained from ECMWF (Kaiser et al., 2012).

MASINGAR runs coupled ("inline") with an atmospheric general circulation model, MRI-AGCM3 (Yukimoto et al., 2012), which provides meteorological variables (horizontal winds and air temperature) and ground properties (surface temperature, soil moisture, snow cover, etc.). The meteorological variables (horizontal winds and air temperature) are constrained by JMA's operational global analysis (GANAL) (Japan Meteorological Agency, 2002) and global forecast at 6-hour interval by Newtonian nudging. The JMA Global Merged SST analysis is used for the sea surface temperature. MASINGAR has been employed by JMA for operational dust prediction since 2004, and is also used for climate research (e.g., CMIP6 and CCMI) as a part of the climate projection model MRI-CGCM3 (Meteorological Research Institute coupled general circulation model version 3) (Yukimoto et al., 2012). The model also contributes to the WMO

Sand and Dust Storm Warning Advisory and Assessment System for Asia (http://eng.nmc.cn/sds_was.asian_rc/).

Two types of aerosol data assimilation systems are available in MASINGAR. The variational-based system (MASINGAR/2D-Var; Yumimoto et al. 2018) was used in the development of the JRAero aerosol reanalysis product (Yumimoto et al. 2017) and scheduled for operational use for dust prediction using the AOT from the geostationary satellite Himawari-8. The ensemble-based system (MASINGAR/LETKF; Sekiyama et al., 2016; Yumimoto et al., 2016a) was developed as a research version and applied to assimilation experiments with both a space-based Lidar and Himawari-8 inputs (Sekiyama et al., 2010; Yumimoto et al., 2016b).

For the ICAP-MME, the model resolution was upgraded from T106L30 Gaussian grid (approximately 110 km with 30 vertical layers in the hybrid sigma-pressure levels from the surface to 0.4 hPa) to TL319L40 grid (approx. 60 km with 40 vertical layers from the surface to 0.4 hPa) in 2013. The horizontal grid resolution was further enhanced to T479 (approx. 40 km) in February 2017. A quality controlled AOT from MODIS NRT L3 product (MCDAODHD) (Zhang and Reid, 2006) has been assimilated every 6 hours in the variational-based system since August 2016.

A4. FMI SILAM

The System for Integrated modeLling of Atmopspheric composition (SILAM) (http://silam.fmi.fi) has been developed in FMI for operational (since 2001) and research calculations of atmospheric composition at regional to global scale. SILAM has two transport cores -- Lagrangian particle model (Sofiev et al., 2006) and Eulerian (Sofiev et al., 2015). The Eulerian transport scheme used in all atmospheric composition simulations is combined with an

adaptive vertical diffusion algorithm (Sofiev, 2002). A detailed aerosol dry deposition scheme of (Kouznetsov and Sofiev, 2012) is accompanied with the gaseous surface uptake scheme based on the resistance analogy approach. For secondary inorganic aerosol formation, the chemistry scheme of the DMAT model (Sofiev, 2000) is extended with the coarse-nitrate formation in marine boundary layer. Dynamic emission schemes have been developed for sea-salt (Sofiev et al., 2011), wild-land fires IS4FIRES system v.2, (Soares et al., 2015; Sofiev et al., 2009), wind-blown dust following the modified saltation approach (Marticorena and Bergametti, 1995; Zender, 2003), and biogenic VOC emission after (Poupkou et al., 2010).

Being an offline model SILAM has an interface to widely used sources of meteorological information, such as ECMWF, HIRLAM, HARMONIE and WRF models, as well as to the GCM systems, such as ECHAM and NorESM. The model includes a meteorological pre-processor for ensuring the solenoidal wind flow and for diagnosing the basic features of the boundary layer and the free troposphere (such as diffusivities, similarity scales, and latent and sensible heat fluxes) from the input meteorological fields (Sofiev et al., 2010).

SILAM implements several data assimilation techniques for 3D-var, 4D-var, ensemble Kalman filter and ensemble Kalman smoother data-assimilation techniques (Vira et al., 2017; J Vira and Sofiev, 2012; J. Vira and Sofiev, 2012). The model is also capable of stand-alone adjoint simulations for, e.g., sensitivity analysis.

Scales of the SILAM applications vary from gamma-mesoscale up to global with characteristic resolution of 0.1-0.5 degree (Lehtomäki et al., 2018; Sofiev et al., 2018). SILAM is a part of Copernicus Atmospheric Service CAMS-50 (Marécal et al., 2015). The model has been evaluated against air quality observations in Europe and worldwide via both dedicated studies

and within the operational quality assurance procedures (Huijnen et al., 2010), http://www.gmes-atmoshpere.eu, http://www.myair.eu (Solazzo et al., 2012a, 2012b).

A5. Météo-France MOCAGE

MOCAGE (Modèle de Chimie Atmospherique à Grande Echelle) is an off-line chemistry transport model used for research at Météo-France in a wide range of scientific studies on tropospheric and stratospheric chemistry at various spatial and temporal scales. It was used for example for studying the impact of climate on chemistry (Teyssèdre et al., 2007; Lacressonnière et al., 2012; Lamarque et al., 2013) or tropospheric–stratospheric exchanges using data assimilation (El Amraoui et al., 2010;Barré et al., 2013). MOCAGE is also used for daily operational air quality forecasts in the framework of French platform Prev'Air (Rouil et al., 2009, http://www2.prevair.org/) and in the European CAMS (Copernicus Atmospheric Monitoring Services) project by being one of the models contributing to the regional ensemble forecasting system over Europe (Marécal et al., 2015, http://macc-raq-op.meteo.fr/index.php). MOCAGE uses the semi-lagrangian advection scheme from for the grid-scale transport, while the convective transport and the turbulent diffusion are parameterized. Required meteorological fields are taken from operational analysis from the ARPEGE model (Action de Recherche Petite Echelle Grande Echelle) operated at Météo-France (Courtier et al., 1991). MOCAGE includes the RACM scheme for tropospheric chemistry and the REPROBUS scheme for stratospheric chemistry (Stockwell et al., 1997). MOCAGE allows representation of desert dust, sea salt, black carbon, primary organic carbon and secondary inorganic aerosols (sulfate, nitrate ammonium). It uses a sectional representation with 6 bins for each aerosol, ranging from 2nm to 50microns (Sic et al, 2015; Guth et al, 2016).

A6. NASA GEOS-5

The Goddard Earth Observing System model, version 5 (GEOS-5), is a global Earth system model developed at the NASA Global Modeling and Assimilation Office (GMAO, Rienecker et al., 2008, Molod et al. 2015). GEOS-5 serves NASA (1) as a state-of-the-art modeling tool to study climate variability and change, (2) as a provider of research quality reanalyses for use by NASA instrument teams and the scientific community at large, and (3) as a source of near-real time forecasts of aerosol and atmospheric constituents in support of NASA aircraft campaigns (e.g., KORUS-AQ, ORACLES). GEOS-5 includes components for atmospheric circulation and composition (including atmospheric data assimilation), ocean circulation and biogeochemistry, and land surface processes. Components and individual parameterizations within components are coupled under the Earth System Modeling Framework (ESMF, Hill et al., 2004). GEOS-5 has a mature atmospheric data assimilation system that builds upon the Grid-point Statistical Interpolation (GSI) algorithm jointly developed with NCEP (Rienecker et al., 2008) and is currently evolving into a hybrid ensemble-variational assimilation system. The version of GEOS-5 documented here is run in near-real time on a cubed-sphere grid at a nominal 25 km horizontal resolution (output is saved on a 0.25^{o}x0.3125^{o} latitude x longitude grid) with 72 vertical hybrid sigma levels from the surface to approximately 85 km.

In addition to traditional meteorological parameters (winds, temperatures, etc.), GEOS-5 includes modules to represent aerosols and tropospheric–stratospheric chemical constituents, and their respective radiative feedback. Aerosols are handled through a version of the GOCART (Chin et al., 2002; Colarco et al., 2010) run online and radiatively coupled in GEOS-5. GOCART

treats the sources, sinks and chemistry of dust, sulfate, sea salt and black and organic carbon aerosols. Aerosol species are assumed to be external mixtures. Aerosol and precursor emissions in the near-real time system are similar to those in the recent GEOS-5 produced Modern-era Retrospective analysis for Research and Analysis, Version 2 (MERRA-2, Randles et al. 2017). Dust and sea salt have wind speed-dependent emissions and discretize the particle size distribution across five size bins apiece. Total mass of sulfate and hydrophobic and hydrophilic modes of carbonaceous aerosols are tracked. Biomass burning emissions of sulfur dioxide and carbonaceous aerosols are from the Quick Fire Emission Data set (QFED, Darmenov and da Silva, 2013). Aerosol optical property assumptions are as in Randles et al. (2017), including a treatment for non-spherical dust particles (Colarco et al., 2014b). In January 2017 the near-real time GEOS-5 system was updated to include a series of tracers for nitrate aerosols (including three size bins of nitrate) following the methodology in Bian et al. (2017).

The aerosol data assimilation methodology also follows from the description in Randles et al. (2017). In near-real time, GEOS-5 includes assimilation of AOD observations from the MODIS sensors on both Terra and Aqua satellites. Based on the work of Zhang and Reid (2006) and Lary et al. (2010), we originally developed a back-propagation neural network to correct observational biases related to cloud contamination, surface parameterization and aerosol microphysics. This empirical algorithm has been adapted to retrieve AOD directly from cloud-cleared MODIS reflectance. Online quality control is performed with the adaptive buddy check of Dee et al. (2001), with observation and background errors estimated using the maximum likelihood approach of Dee and da Silva (1999). Following a multi-channel AOD analysis, three-dimensional analysis increments are produced exploring the Lagrangian characteristics of the

problem, generating local displacement ensembles intended to represent misplacements of the aerosol plumes.

A7. NOAA NGAC

In March, 2017, NCEP implemented the NEMS GFS Aerosol Component (NGAC) version 2 (NGACv2) multi-species aerosol forecast into operation. The aerosol species include organic carbon aerosols, black carbon aerosols, sea salt and sulfate aerosols and dust. NGACv2 uses an updated atmosphere model Global Forecast System (GFS) implemented in May 2016. The convection scheme is changed to the Relaxed Arakawa–Schubert scheme (the RAS scheme, Moorthi and Suarez, 1992, 1999) due to the need of vertical aerosol transport. The aerosol model is NASA/GSFC's GOCART aerosol module (Colarco et al. 2010). Black carbon and organic carbon aerosols are tracked separately. The organic carbon is presented as particulate organic matter. The chemical processing of carbonaceous aerosols as a conversion from a hydrophobic to hydrophilic mode follows Cooke et al. (1999) and Chin et al. (2002) with an e-folding timescale of 2.5 days (Maria et al. 2004). Following Colarco (2014), five size bins of sea salt aerosol particles with a dry radius range of 0.03-10mm are considered for an indirect production mechanism from bursting bubbles (Monahan et al. 1986), and later modified by Gong (2003). Four sulfate tracers dimethyl sulfide (DMS), sulfur dioxide (SO_2), sulfate (SO4), and methane sulfonic acid (MSA) are tracked. Sulfate chemistry includes the DMS oxidation by hydroxyl radical (OH) during the day and by nitrate radical (NO3) at night to form SO_2, and SO_2 oxidation by OH in the gas phase and by hydrogen peroxide (H_2O_2)in the aqueous phase to form sulfate, as described in Chin et al. (2002). The AOD is computed from the complex refractive indices, size distributions, and the hygroscopic properties of aerosols following Chin et al. A

computational error on dust AOD calculation is fixed, and the removal process has been tuned to improve dust performance.

NGACv2 runs at T126 L64 resolution and provides 5-day multi-species forecasts, twice per day for the 00:00UTC and 12:00UTC cycles. The aerosol initial conditions are taken from the 12 hour NGAC forecast from the previous cycle while meteorological initial conditions are from the down-scaled high-resolution Global Data Assimilation System (GDAS) analysis. NGACv2 provides products in addition to those from NGACv1 dust-related products. Total Aerosol Optical Depth (AOD) and AOD from each species are produced to support global and regional multi-model ensemble aerosol forecasts. Single scattering albedo and asymmetric factor for total aerosols at 340nm are produced to support UV index forecast are available. Besides these fields, the three-dimensional mixing ratios for each aerosol species at model levels are also produced. The data are publicly available at: http://www.nomads.ncep.noaa.gov/pub/data/nccf/com/ngac/prod

A8. UKMO Unified Model

The dust forecasts from the UK Met Office are produced by the global NWP configuration of the Met Office Unified Model (MetUM). The dust scheme is essentially that of Woodward (2001) with modifications as described in Woodward (2011) and Collins et al. (2011). The dust emission scheme is based on Marticorena and Bergametti (1995) and represents an initial horizontal/saltation flux in a number of size bins with subsequent vertical flux of bare soil particles from the surface into the atmosphere. The global NWP model transports only 2 bins (0.1–2 microns and 2–10 microns radii), calculated from the emissions with the original 9 bins using a prescribed size distribution broadly consistent with Kok et al. (2011). The magnitude of

the emission is a cubic function of the exceedance of the friction velocity over bare soil with respect to a threshold value, where this friction velocity is determined from the model wind field and boundary layer structure, and the threshold friction velocity is increased by the presence of soil moisture according to Fécan (1999). The conversion from the horizontal flux to the vertical flux is first limited using the clay fraction in the soil texture data set, according to Gillette (1978), and then partitioned into the new bins by prescribing the emitted size distribution. Once the dust is lifted into the atmosphere it is transported as a set of tracers by the model 3-D wind field. Johnson et al. (2011) gave in-depth description and evaluation of the Met Office dust forecasts, in a local area model over North Africa. Dust is assimilated in a 4-D Var framework following Benedetti et al. (2009), using aerosol observations from MODIS on-board NASA's Aqua platform. Initially, MODIS (Collection 5.1) observations (best quality, dust filtered) were assimilated only over the land based on MODIS Dark Target (Kaufman et al., 1997a, b; Levy et al., 2007, 2009) and Deep Blue (Hsu et al., 2004, 2006) retrievals. Incremental updates to the operational system includes MODIS observation over ocean in Feb 2015, and an updated version of MODIS AOD (Collection 6.0) in Dec 2017.

A9. US NAVY NAAPS

The Navy Aerosol Analysis and Prediction System (NAAPS) is the US Navy's global aerosol forecast model, which produced the world's first operational global aerosol forecasts and then was the first with aerosol data assimilation. In its current operational configuration, NAAPS makes 6-day forecasts, 4 times a day at 1080x540 global (1/3 degree) spatial resolution and 35 vertical levels driven by the Navy Global Environmental Model (NAVGEM) meteorology (Hogan et al., 2014). It has multiple research versions, including the NAAPS reanalysis (Lynch

et al., 2016) and ensemble NAAPS (Rubin et al, 2016). Quality controlled retrievals of AOD from the Moderate Resolution Imaging Spectroradiometer (MODIS) (Zhang et al., 2006; Hyer et al., 2011; Shi et al., 2014) are assimilated through the Navy Atmospheric Variational Data Assimilation System (NAVDAS) for AOD (NAVDAS-AOD; Zhang et al., 2008) in the operational run, while the model has capabilities of assimilating other quality controlled observations, including lidar back scatter vertical profiles and AOD products from other platforms through either variational or ensemble methods (Zhang et al., 2011; Rubin et al., 2017; Hyer et al., 2018).

NAAPS characterizes anthropogenic and biogenic fine (ABF, including sulfate, and primary and secondary organic aerosols), dust, biomass burning smoke and sea salt aerosols. A 1st order approximation of secondary organic aerosol (SOA) processes is adopted in which production of SOA from its precursors is assumed to be instantaneous and included with the sulfate species to form a combined anthropogenic and biogenic fine (ABF) species (Lynch et al, 2016). Smoke from biomass burning is derived from near-real time satellite based thermal anomaly data used to construct smoke source functions with regional corrections (Reid et al., 2009). Dust is emitted dynamically and is a function of modeled friction velocity to the fourth power, surface wetness and surface erodibility (Westphal et al., 1988). Sea salt emission is driven dynamically by sea surface wind, according to the parameterization described by Witek et al. (2007).

Appendix B: Definition of terminologies

Root Mean Square Error (RMSE):

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=0}^{n}(\tau_{model} - \tau_{obs})_i^2}$$

where τ represents AOD, and n is the total number of observational or model data.

Bias: $\tau_{model} - \tau_{obs}$

Mean error: $\frac{1}{n}\sum_{i=1}^{n}(\tau_{model} - \tau_{obs})_i$

Mean absolute error: $\frac{1}{n}\sum_{i=1}^{n}|\tau_{model} - \tau_{obs}|_i$

Coefficient of determination: $r^2 = \frac{\left(\sum_{i=1}^{n}(x_i-\overline{x})(y_i-\overline{y})\right)^2}{\sum_{i=1}^{n}(x_i-\overline{x})(y_i-\overline{y})\sum_{i=1}^{n}(x_i-\overline{x})(y_i-\overline{y})}$

where $\overline{x}$ and $\overline{y}$ are the mean values of variable x and y.

Absolute Forecast Error (AFE): $|\tau_{model} - \tau_{obs}|$ for forecast mode.

Ensemble mean: $\frac{1}{m}\sum_{i=1}^{m}x_i$ where m is the total number of the individual models.

Ensemble spread is defined as the standard deviation of all the individual models, ie.,

$$\sigma = \sqrt{\frac{1}{m}\sum_{i=1}^{m}(x_i - \overline{x})^2}$$

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ICAP Aerosol Multi-Model Ensemble

Table 1. Basic properties and configurations of ICAP input models as of June 2017.

Model	MONARCH (former BSC-CTM)	CAMS/IFS	MASINGAR	SILAM	MOCAGE	GEOS-5	NGAC	MetUM	NAAPS
Organization	BSC	ECMWF/CAMS (former MACC)	JMA	FMI	Meteo France	NASA	NOAA	UKMO	US NAVY
Status	QO	O	QO	O	O	QO	O	O	O
Meteorology	Inline NMMB	Inline IFS	Inline AGCM	Offline IFS	Offline ARPEGE	Inline GEOS-5	Inline GFS	Inline UM	Offline NAVGEM
Resolution (deg lat x lon)	1.4x1 (0.7x0.5)[p]	0.35x0.35	0.38x0.38	0.5x05	1x1	0.25x0.31	1x1	0.35x0.23	0.35x0.35
Levels	24 (48) [p]	60	40	60	47	72	64	70	35
DA	LETKF[p]	4DVar	2DVar LETKF[p]	3Dvar[p], 4Dvar[p], EnKF[p]	NA	2DVar+LDE	NA	4DVar	2DVar 3DVar[p], EnKF[p]
Assimilated Aerosol Observation	(DAQ MODIS DT+DB)[p]	DAQ MODIS DT+DB PMAp	MODIS L3 AHI[p] CALIOP[p]	NA	NA for this release	Neural Net MODIS	NA	MODIS Dust AOD	DAQ MODIS L3, VIIRS[p] CALIOP[p]
Species	Dust, SS, (BC, OC, Sulfate)[p]	BC, Dust, OC, SS, Sulfate	BC, Dust, OC, SS, Sulfate	BC, Dust, OC, SS, Sulfate, Nitrate, B. Burning Smoke	BC, Dust, OC, SS, Sulfate, Nitrate, Ammon.	BC, Dust, OC, SS, Sulfate, Nitrate*	BC, Dust, OC, SS, Sulfate	Dust	Anthro+Bio. Fine, B. Burn. Smoke, Dust, SS
Size Bins	8 (dust, SS), bulk for others[p]	3 (dust, SS), bulk for others	10 (dust, SS), bulk for others	4 (dust), 5 (SS), 3 (B. Burning smoke), 2 (sulfate), bulk for others	6	5 (dust, SS), 2 (BC, OC), 3 (NI*), bulk sulfate	5 (dust, SS), 2(BC, OC), bulk sulfate	2	Bulk
Anthro. and biogenic Emissions	NA	MACCity (anthro), MEGAN (biogenic)	MACCity	MACCity, STEAM, MEGANE, HTAP (Coarse PM)	MACCity (anthro.) MEGAN-MACC (biogenic)	EDGAR V4.1/4.2, AeroCom Phase II, GEIA	EDGAR V4.1, AeroCom Phase II, GEIA	NA	MACCity, BOND, POET
Bio. Burn. Emissions	NA (GFAS)[p]	GFAS	GFAS	GFAS, IS4FIRES	MACCity	QFED	GBBEPx	NA	FLAMBE

Note: "p" means prototype, and not in operational mode. "O" stands for operational and "QO" for quasi-operational. Species "SS" presents sea salt. *Nitrate aerosols were added to GEOS-5 near-real time system as of January 24, 2017.

Table 2. For each of the 21 AERONET sites used in this study, AERONET Version3 Level1.5 mean total AOD at 550nm and root mean square error (RMSE) for 6-hr and 72-hr forecasts from the four core ICAP models and the ICAP-MME for June 2016-May 2017.   RMSE values are listed in order from low to high, and ICAP-MME results are shown in boldface. Sample size refers to the number of valid 6-hour average AERONET AOD observations over this 12-month time period.

site	Location	Main Aerosol type	Mean Aeronet total AOD	6-hr forecast RMSE					72-hr forecast RMSE					sample size
Alta Floresta	Brazil, 9S, 56W	smoke	0.21	**0.11**	0.13	0.13	0.14	0.21	**0.12**	0.14	0.14	0.15	0.16	248
Amsterdam Island	Southern Indian Ocean, 38S, 78E	Sea salt	0.07	0.04	0.04	**0.05**	0.07	0.08	0.04	0.04	**0.05**	0.06	0.09	273
Banizoumbou	Sahel, 13N, 2E	Dust	0.49	**0.21**	0.22	0.23	0.29	0.29	0.24	**0.24**	0.27	0.30	0.33	641
Beijing	China, 39N, 116E	ABF, dust	0.54	**0.41**	0.41	0.42	0.47	0.48	**0.46**	0.46	0.49	0.52	0.56	451
Cape Verde	Sub-tro. Atlantic, 16N, 22W	Dust	0.42	0.14	**0.14**	0.16	0.19	0.22	0.18	**0.19**	0.21	0.25	0.26	393
Cart Site	Great Plains, 36N, 97W	Clean	0.09	0.05	**0.06**	0.06	0.07	0.09	0.05	**0.06**	0.06	0.08	0.10	612
Chapais	Quebec, 49N, 74W	Clean	0.07	0.03	**0.04**	0.04	0.04	0.07	**0.04**	0.04	0.05	0.05	0.06	245
Chiang Mai	Thailand, 18N, 98E	smoke	0.42	0.20	**0.22**	0.22	0.26	0.29	0.23	**0.24**	0.25	0.29	0.31	295
Gandhi College	Rural India, 25N, 84E	Dust, pollution	0.62	**0.23**	0.23	0.24	0.25	0.29	**0.27**	0.27	0.28	0.31	0.33	308
GSFC	E. CONUS, 38N, 76W	Pollution	0.10	0.05	0.05	**0.06**	0.07	0.09	**0.07**	0.07	0.07	0.09	0.11	614
Ilorin	Sahel, 8E, 4E	Smoke, dust	0.73	0.33	**0.33**	0.34	0.37	0.43	0.32	**0.39**	0.39	0.47	0.51	407
Kanpur	Urban India, 26N, 80E	Pollution, dust	0.62	**0.29**	0.30	0.31	0.32	0.32	0.37	**0.39**	0.39	0.44	0.45	655
Minsk	Western Asia, 53N, 27E	Pollution, smoke	0.13	0.06	**0.07**	0.07	0.08	0.10	0.07	**0.09**	0.11	0.11	0.14	397
Moldova	Eastern Europe, 47N, 28E	Pollution	0.17	**0.09**	0.11	0.11	0.11	0.12	**0.10**	0.11	0.13	0.13	0.15	383
Monterey	W. CONUS, 36N, 121W	Clean	0.27	0.27	**0.46**	0.46	0.56	1.01	0.28	0.56	**0.58**	0.59	1.01	148
Palma de Mallorca	Mediterranean, 39N, 2E	Dust, ABF	0.12	**0.05**	0.05	0.05	0.07	0.08	**0.08**	0.08	0.08	0.08	0.12	767
Ragged Point	Caribbean, 13N, 59W	African dust	0.17	0.06	**0.06**	0.07	0.08	0.10	**0.08**	0.08	0.09	0.10	0.12	416
Rio Branco	Brazil, 9S, 67W	smoke	0.23	0.15	**0.15**	0.19	0.20	0.21	**0.18**	0.20	0.20	0.22	0.25	232
Singapore	Maritime Cont., 1N, 103E	ABF, smoke	0.28	0.19	**0.20**	0.22	0.22	0.22	**0.22**	0.23	0.23	0.25	0.25	410
Mezaira	Southwest Asia, 23N, 54E	Dust	0.36	**0.15**	0.16	0.18	0.21	0.24	0.18	**0.19**	0.23	0.24	0.43	641
Yonsei University	South Korea, 38N, 127E	Dust, Pollution	0.45	**0.25**	0.26	0.26	0.28	0.30	**0.30**	0.30	0.32	0.33	0.33	523

Table 3. AERONET Version3 Level1.5 mean coarse-mode AOD at 550 nm, and ICAP models root mean square error (RMSE) from their 6-hr and 72-hr forecasts for June 2016-May 2017 for dust-influenced sites. The RMSEs for the 8 ICAP members and ICAP-MME (bold) are listed sequentially from low to high for each site.

site	Mean AERONET coarse AOD	6-hr dust forecast RMSE								72-hr dust forecast RMSE							
Banizoumbou	0.37	0.18	0.18	**0.19**	0.22	0.23	0.27	0.29	0.45	0.21	0.22	**0.22**	0.22	0.26	0.28	0.34	0.47
Beijing	0.15	**0.12**	0.12	0.13	0.15	0.15	0.15	0.20	0.29	0.12	**0.13**	0.13	0.15	0.16	0.17	0.22	0.36
Capo Verde	0.33	**0.13**	0.13	0.14	0.14	0.17	0.21	0.21	0.22	0.16	**0.16**	0.18	0.19	0.22	0.23	0.23	0.25
Gandhi College	0.22	0.11	0.11	0.11	**0.13**	0.14	0.14	0.17	0.21	0.10	0.10	0.11	**0.13**	0.15	0.15	0.17	0.22
Ilorin	0.42	**0.17**	0.20	0.20	0.21	0.24	0.24	0.26	0.30	**0.17**	0.20	0.20	0.24	0.24	0.29	0.30	0.34
Kanpur	0.22	**0.11**	0.12	0.12	0.13	0.13	0.15	0.16	0.19	0.10	**0.13**	0.14	0.14	0.14	0.15	0.16	0.20
Palma de Mallorca	0.06	**0.04**	0.04	0.05	0.05	0.06	0.06	0.06	0.09	**0.06**	0.06	0.07	0.07	0.07	0.07	0.10	0.10
Ragged Point	0.14	0.07	**0.08**	0.08	0.08	0.10	0.10	0.11	0.11	0.07	**0.09**	0.10	0.10	0.10	0.10	0.12	0.12
Mezaira	0.22	0.11	0.13	**0.13**	0.14	0.15	0.21	0.25	0.31	0.12	0.15	0.15	**0.16**	0.22	0.23	0.36	0.42
Yonsei University	0.12	0.07	0.08	**0.08**	0.10	0.10	0.11	0.11	0.12	0.07	0.07	**0.08**	0.10	0.11	0.12	0.12	0.15

Table 4. Coefficient of determination (r^2) between 2 out of 3 variables: ensemble mean, ensemble spread and absolute forecast error (AFE) of the ICAP MME for dust AOD 6-hr and 72-hr forecasts at the dusty AERONET sites. All calculations are based on model and AERONET V3 Level1.5 data during Jun 2016-May 2017. Value less than 0.04 means the correlation is not statistically significant at 95% level by Student-t test.

site	6-hr dust fcst			72-hr dust fcst			
	ensemble mean & AFE	ensemble spread & AFE	ensemble mean & spread	ensemble mean & AFE	ensemble spread & AFE	ensemble mean & spread	sample size
Banizoumbou	0.13	0.08	0.52	0.10	0.06	0.60	647
Beijing	0.13	0.12	0.86	0.16	0.14	0.83	451
Capo Verde	0.19	0.15	0.47	0.12	0.07	0.61	401
Gandhi College	0.05	0.03	0.88	0.06	0.04	0.87	315
Ilorin	0.17	0.09	0.70	0.24	0.16	0.82	409
Kanpur	0.04	0.01	0.59	0.01	0.00	0.68	663
Palma de Mallorca	0.27	0.20	0.83	0.48	0.38	0.79	776
Ragged Point	0.11	0.11	0.83	0.13	0.10	0.80	420
Mezaira	0.54	0.46	0.70	0.57	0.45	0.59	649
Yonsei University	0.03	0.02	0.79	0.03	0.02	0.83	530

Table 5. Estimated forecast error for total AOD based on linear regressions of absolute forecast errors (AFE) on ensemble mean, ensemble spread and both respectively for forecasts with different forecast hours. Coefficient of determination (r^2) between the predicted forecast errors using the equations below and forecast error of the ICAP MME consensus is also listed. All the r^2 values here are statistically significant at 95% level by student-t test (criteria of 0.04).

forecast hour	Total AOD forecast error estimate	r^2
6-hr	AFE=0.00+0.36(+-0.01)*mean	0.27
	AFE=0.06+0.59(+-0.01)*spread	0.17
	AFE=0.00+0.31(+-0.01)*mean+0.14(+-0.02)*spread	0.27
24-hr	AFE=0.01+0.43(+-0.01)*mean	0.26
	AFE=0.07+0.79(+-0.02)*spread	0.22
	AFE=0.02+0.30(+-0.01)*mean+0.36(+-0.02)*spread	0.28
48-hr	AFE=0.02+0.47(+-0.01)*mean	0.25
	AFE=0.09+0.74(+-0.02)*spread	0.20
	AFE=0.03+0.35(+-0.01)*mean+0.28(+-0.02)*spread	0.26
72-hr	AFE=0.01+0.43(+-0.01)*mean	0.23
	AFE=0.07+0.61(+-0.01*spread	0.16
	AFE=0.01+0.37(+-0.01)*mean+0.12(+-0.02)*spread	0.23
96-hr	AFE=0.01+0.45(+-0.01)*mean	0.22
	AFE=0.07+0.63(+-0.02)*spread	0.16
	AFE=0.02+0.38(+-0.01)*mean+0.16(+-0.02)*spread	0.23

Table 6. Same as Table 6, except for dust AOD forecast.

forecast hour	Dust AOD forecast error estimate	r^2
6-hr	AFE=0.04+0.23(+-0.01)*mean	0.24
	AFE=0.04+0.45(+-0.01)*spread	0.20
	AFE=0.03+0.18(+-0.01)*mean+0.14(+-0.02)*spread	0.24
24-hr	AFE=0.04+0.28(+-0.01)*mean	0.24
	AFE=0.05+0.52(+-0.01)*spread	0.19
	AFE=0.04+0.24(+-0.01)*mean+0.10(+-0.03)*spread	0.24
48-hr	AFE=0.04+0.35(+-0.01)*mean	0.24
	AFE=0.05+0.61(+-0.02)*spread	0.23
	AFE=0.04+0.30(+-0.01)*mean+0.13(+-0.03)*spread	0.29
72-hr	AFE=0.04+0.28(+-0.01)*mean	0.26
	AFE=0.05+0.44(+-0.01)*spread	0.21
	AFE=0.04+0.23(+-0.01)*mean+0.10(+-0.02)*spread	0.26
96-hr	AFE=0.04+0.27(+-0.01)*mean	0.26
	AFE=0.05+0.43(+-0.01)*spread	0.22
	AFE=0.04+0.22(+-0.01)*mean+0.11(+-0.02)*spread	0.26

Table 7. Contribution of each individual model to the ICAP MME in the 6-hr and 72-hr forecasts for dust AOD at 550 nm. One year mean (June 2016-July 2017) absolute forecast errors from the ICAP MME are listed, as well as those from ensembles based on 6 of 7 dust models with Model 1 (M1), Model 2 (M2), Model 3 (M3), Model 4 (M4), Model 5 (M5), Model 6 (M6) and Model 7 (M7) removed from the ensembles respectively. Percent change of mean absolute forecast error resulting from the removal of each individual model at each site and the average change of all sites are also shown.

site	Dust 6hr fcst error	w/out M1		w/out M2		w/out M3		w/out M4		w/out M5		w/out M6		w/out M7	
Banizoumbou	0.14	0.16	8%	0.15	5%	0.15	1%	0.15	6%	0.14	-1%	0.12	-15%	0.15	4%
Beijing	0.08	0.07	-3%	0.08	1%	0.07	-4%	0.07	0%	0.08	5%	0.08	1%	0.08	10%
Capo Verde	0.08	0.09	4%	0.09	7%	0.09	4%	0.09	2%	0.08	1%	0.08	-7%	0.09	3%
Gandhi College	0.11	0.11	-1%	0.11	4%	0.11	-1%	0.10	-5%	0.11	3%	0.10	-8%	0.12	12%
Ilorin	0.10	0.10	2%	0.10	4%	0.10	-1%	0.10	0%	0.10	3%	0.10	2%	0.10	6%
Kanpur	0.09	0.09	1%	0.09	2%	0.09	-2%	0.08	-5%	0.09	1%	0.08	-9%	0.10	18%
Palma de Mallorca	0.03	0.03	-2%	0.03	3%	0.03	3%	0.03	-1%	0.03	5%	0.03	0%	0.03	10%
Ragged Point	0.05	0.05	0%	0.05	2%	0.05	-3%	0.05	-4%	0.05	0%	0.05	-3%	0.06	15%
Mezaira	0.09	0.09	1%	0.10	4%	0.09	0%	0.09	1%	0.10	12%	0.08	-13%	0.09	2%
Yonsei University	0.06	0.06	-2%	0.06	-3%	0.06	0%	0.06	-4%	0.07	10%	0.06	-5%	0.07	10%
average Dust 6hr-fcst			1%		3%		0%		-1%		4%		-6%		9%
average Dust 72hr-fcst			-1%		2%		0%		2%		6%		-2%		5%
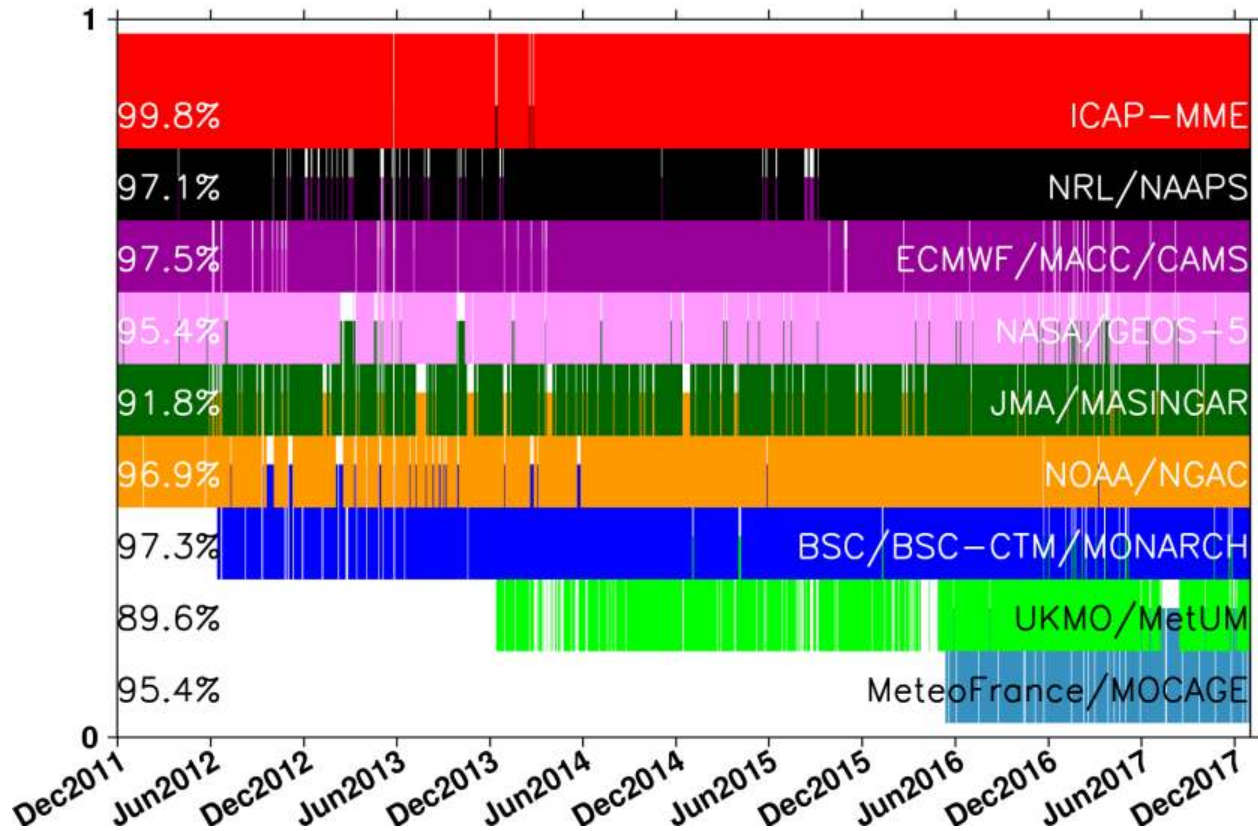
Figure 1. Data availability of ICAP models and ICAP-MME between December 2011 and January 2018. FMI/SILAM model is not shown here because it is a relatively new member and is currently being incorporated into ICAP.
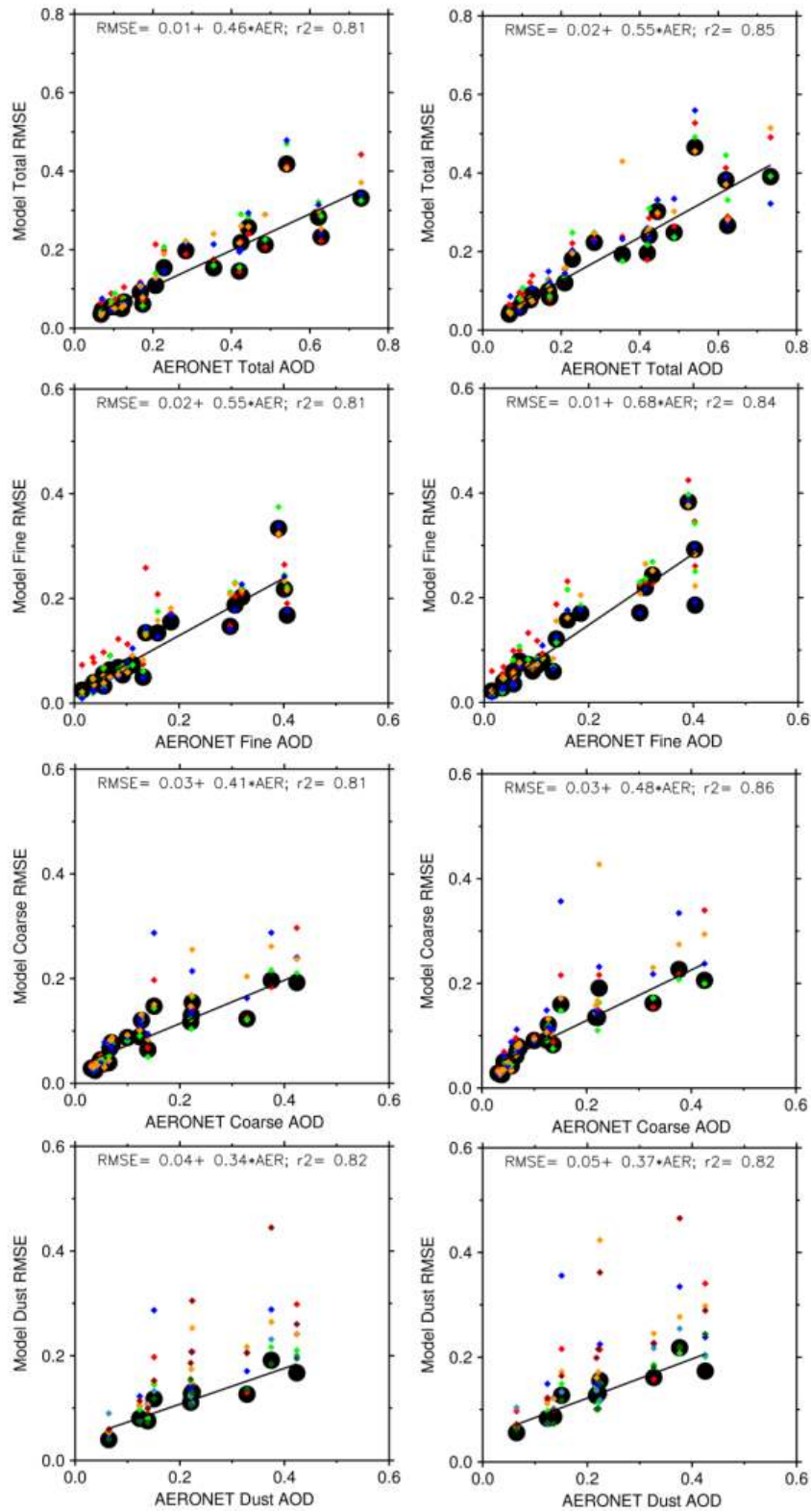
Figure 2. ICAP model 550 nm total, fine, coarse, and dust AOD RMSE versus corresponding

mean AODs for AERONET sites listed in Table 2. Verification of the 6-hr forecasts are on the

left, and the 72-hr forecasts on the right. Large black dots are ICAP-MME consensus means. Individual models are in small colored dots. Validation of dust AOD is based on AERONET coarse-mode AOD at dusty sites listed in Table 3. Monterey site is excluded in the total and fine AOD validation/regression as it is an outlier with its anomalously high and variant biomass-burning smoke levels that resulted from a months-long local wildfire event in the Fall of 2016.
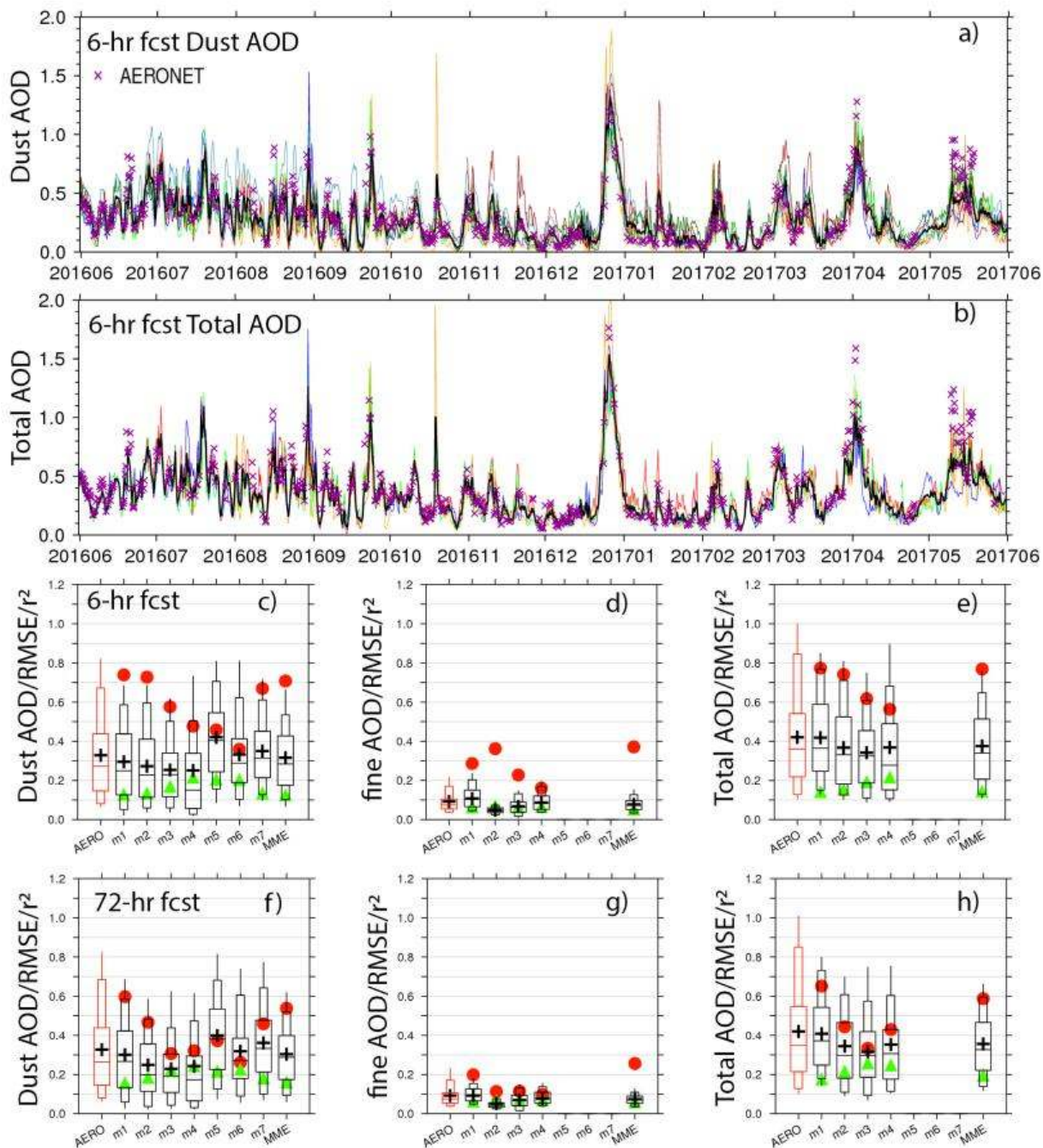
Figure 3. Comparison of ICAP models and AERONET V3 L1.5 550nm AODs at 550 nm for

Capo Verde, an African-dust-influenced site off the west coast of Africa. Included are (a) dust

AOD from the 6-hr forecasts of ICAP models (individual members in colors and ICAP-MME in

black) and the coarse-mode AERONET AOD; b) same as a) except for total AOD. c-d-e) percentiles (95, 90, 75, 50, 25, 10 and 5th) of Dust/Fine/Total AODs of the ICAP model 6-hr forecasts and the paired AERONET data (paired with ICAP-MME, in red). Also shown are the mean model/AERONET AOD values (black plus), RMSE of each model (green triangles) and the coefficient of determination (r^2, red dots) against AERONET observations. f-g-h) same as c-d-e) except for the 72-hr forecasts. All available model data is used in the time series plots, and only paired (paired with AERONET) data is used in the histograms.

a) 6-hr fcst Coarse-mode AOD
   × AERONET

b) 6-hr fcst Fine-mode AOD

c) 6-hr fcst

d)

e)

f) 72-hr fcst

g)

h)

i) TERRA true color

j) TERRA AOD
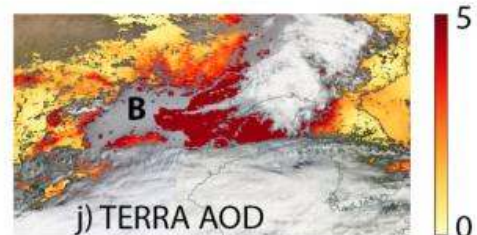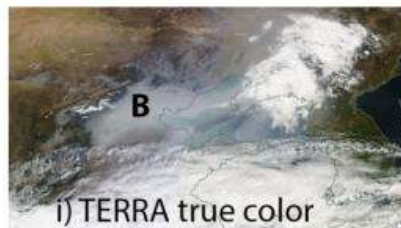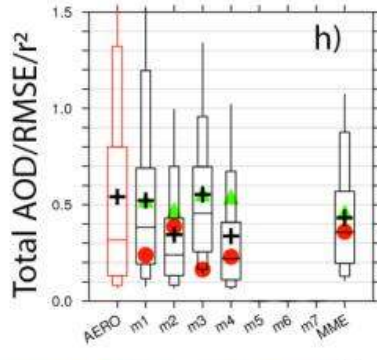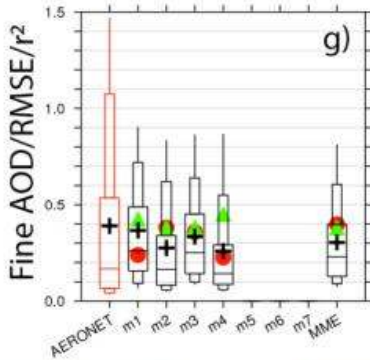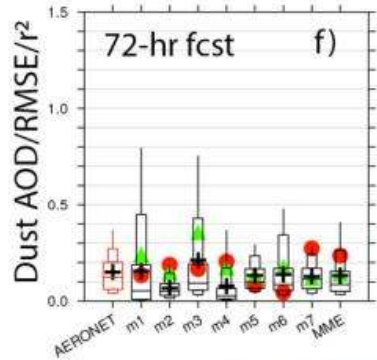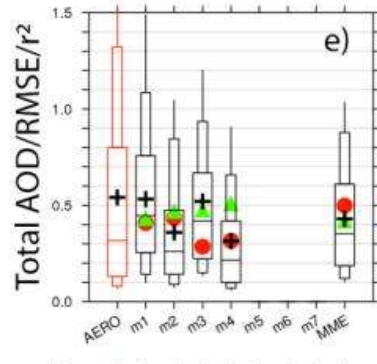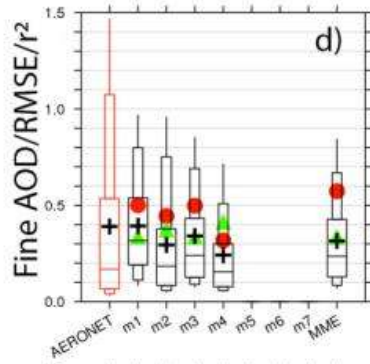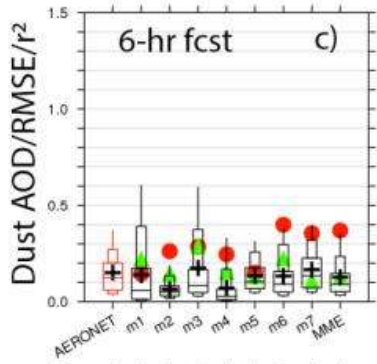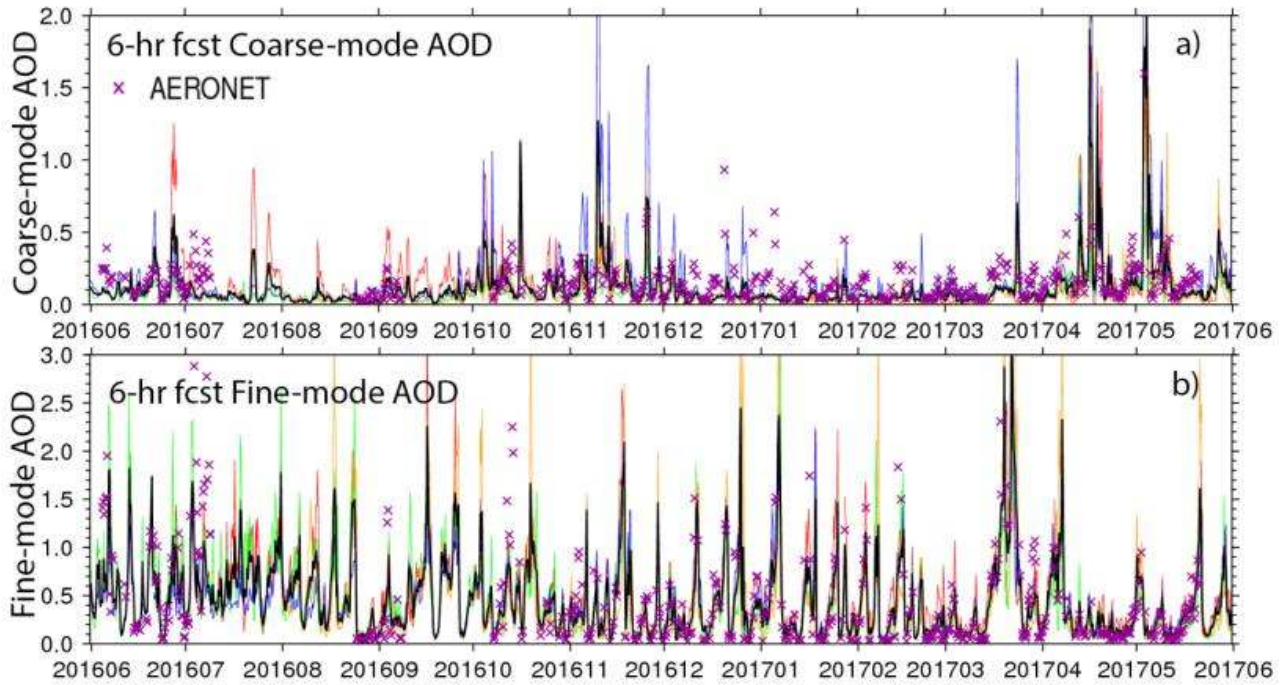
Figure 4. Comparison of ICAP models and AERONET V3 L1.5 AODs at 550 nm for Beijing. Included are (a) dust AOD from the 6-hr forecasts of ICAP models (individual members in colors and ICAP-MME in black) and the coarse-mode AERONET AOD; b) same as a) except for fine-mode AOD. c-d-e) percentiles (95, 90, 75, 50, 25, 10 and 5%) of Dust/Fine/Total AODs of the ICAP model 6-hr forecasts and the AERONET data (paired with ICAP-MME, in red). Also shown are the model mean AOD (black plus), RMSE of each model (green triangles) and the coefficient of determination (r^2, red dots) against AERONET observations. f-g-h) same as c-d-e) except for the 72-hr forecasts. i, j) Terra true color image and AOD respectively for October 13, 2016, with "B" marking the location of Beijing.
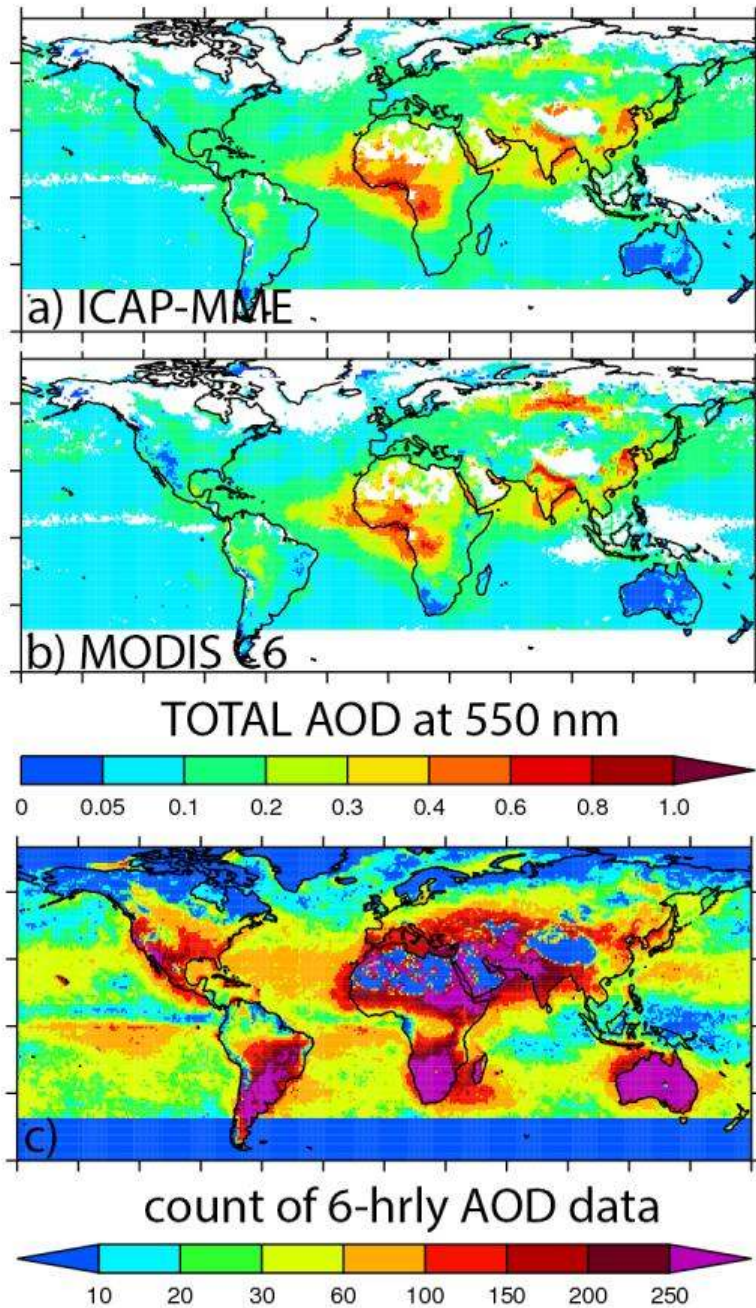
Figure 5. Mean total AOD at 550 nm averaged between Jun 2016 and May 2017 for (a) ICAP-MME 6-hr forecast (spatially and temporally sampled to match MODIS DA-quality data), and (b) DA-quality MODIS C6, and (c) the total number of 6-hrly DA-quality MODIS AOD data. In (a) and (b), only area with DA-quality MODIS data count greater than 15 is shown.
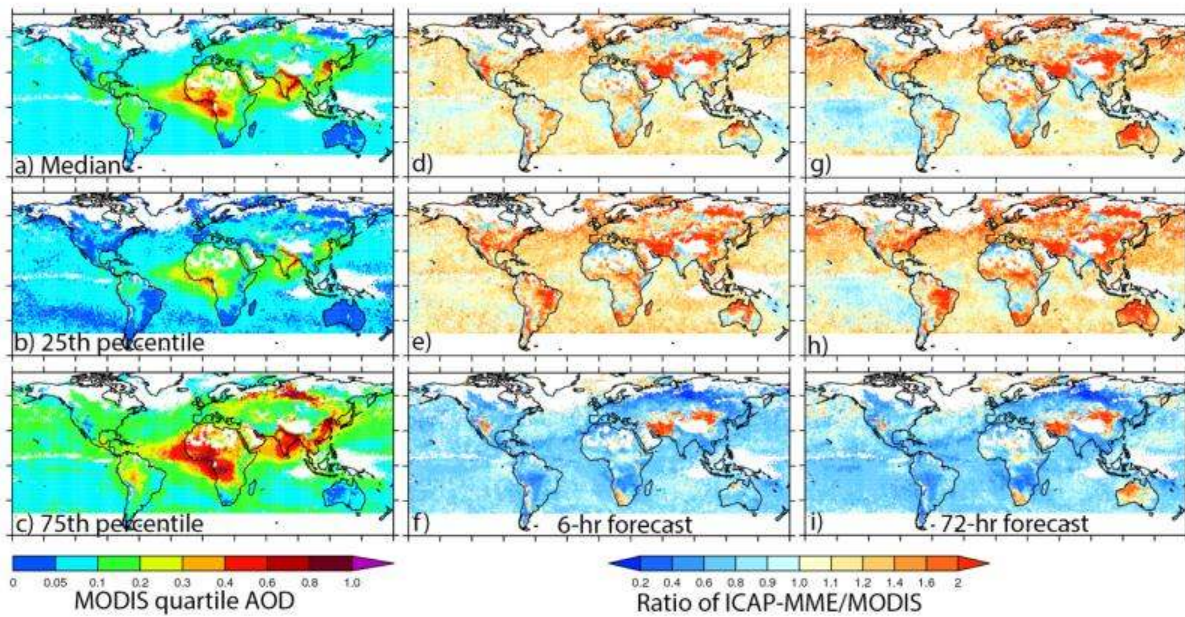
Figure 6. DA-quality MODIS C6 median, 25th and 75th percentiles of total AOD at 550 nm for Jun 2016 – May 2017 time period, and ratios of these quantities from the ICAP-MME 6-hr and 72-hr forecasts to MODIS. ICAP-MME data is spatially and temporally sampled to match MODIS C6 data. Same as Fig. 5, only area with DA-quality MODIS data count greater than 15 is shown.
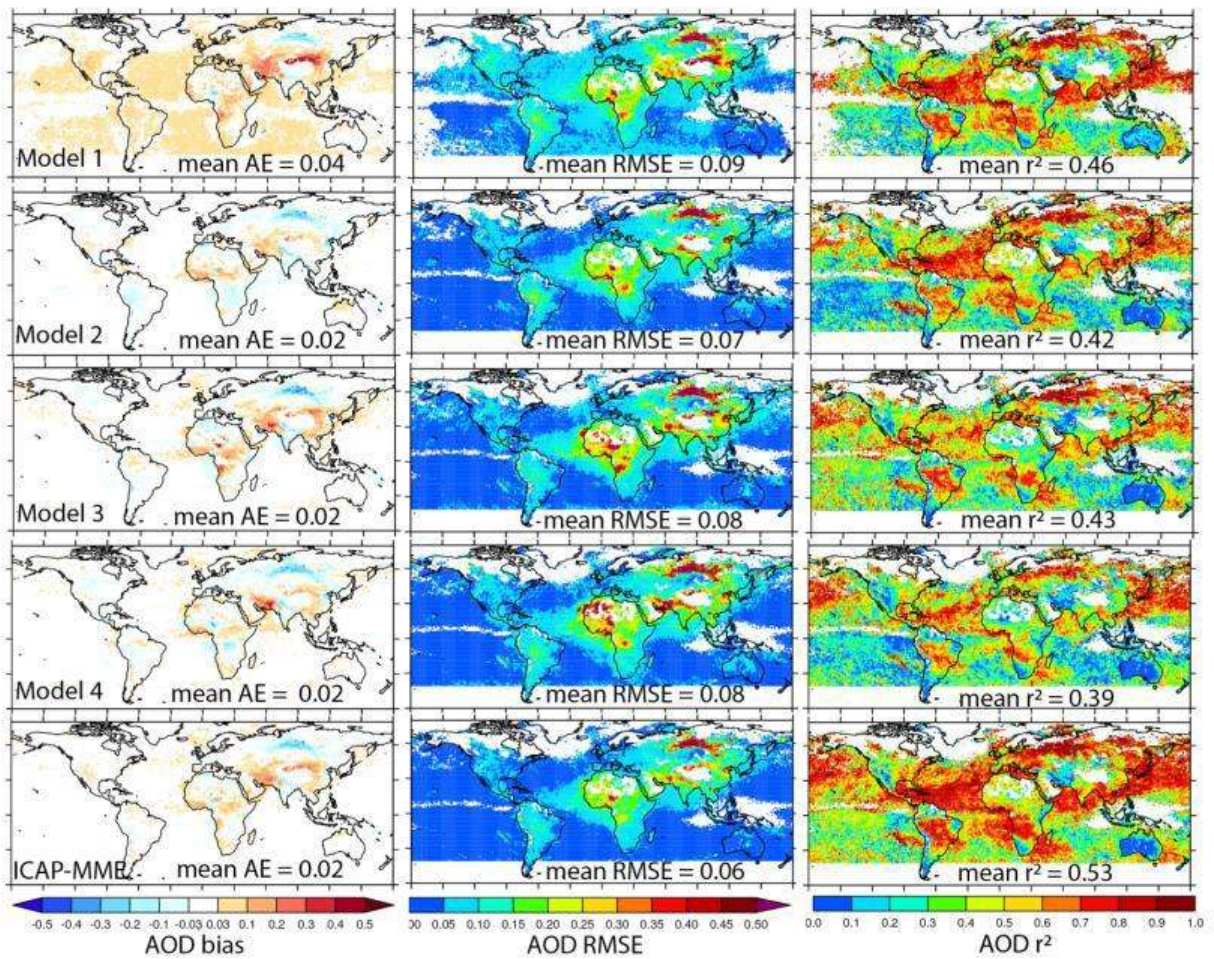
Figure 7. 550 nm mean AOD bias, RMSE and coefficient of determination (r^2) of the 6-hr forecasts (initialized with 00Z analysis) from the four ICAP core models and the ICAP-MME verified against the DA-quality MODIS C6 data for Jun 2016 – May 2017 time period. The numbers inside AOD bias, RMSE and r^2 plots are global mean absolute error (AE), RMSE and r^2. Same as Fig. 5 and 6, statistics are only calculated for grids with DA-quality MODIS data count greater than 15.
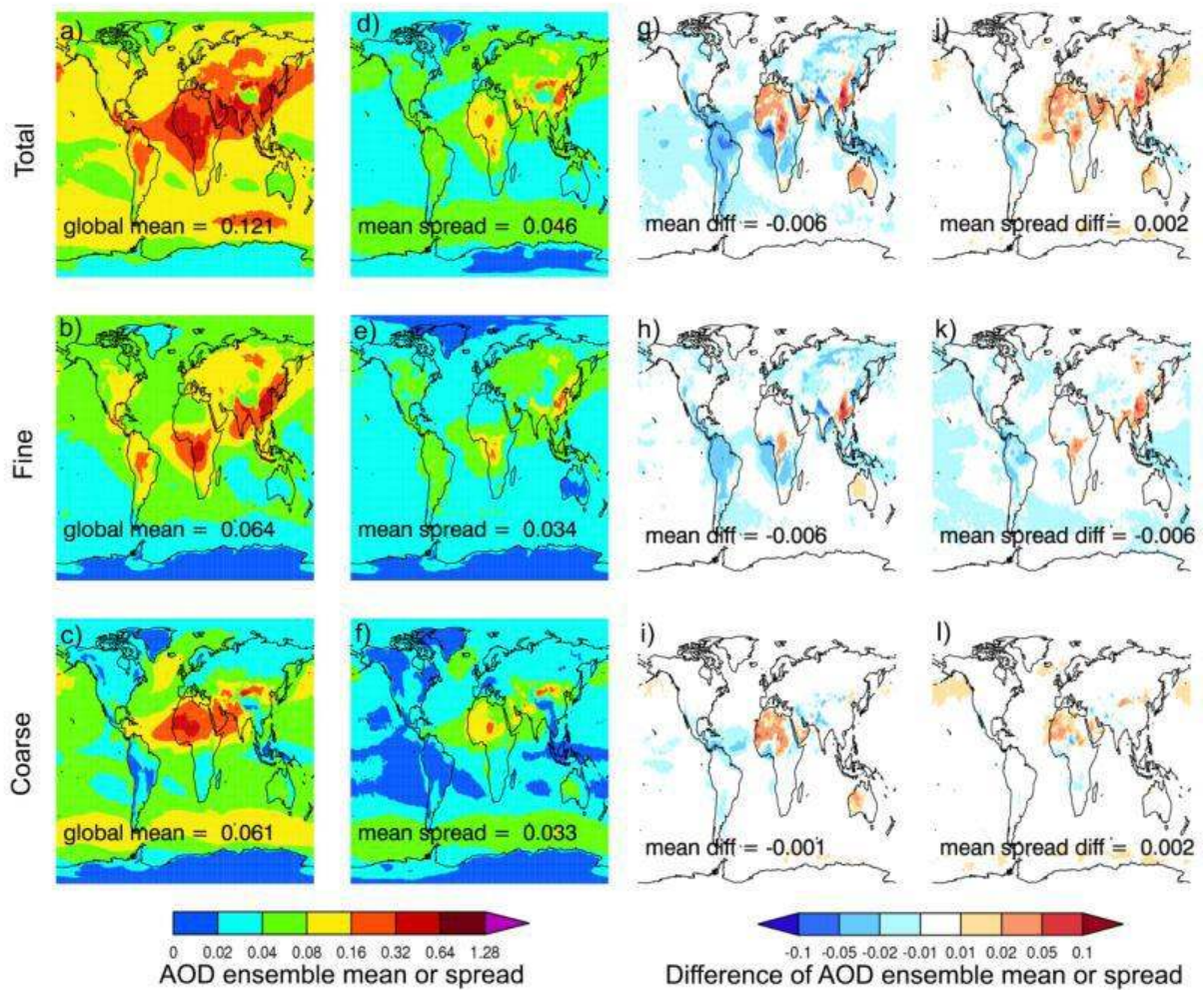
Figure 8. Average ensemble mean and spread of total, fine and coarse-mode AODs at 550 nm among the four core ICAP models for their analysis (approximated with the 6-hr forecasts) mode and the difference relative to the forecast mode (using the 72-hr forecasts) for June 2016-May 2017. The two columns on the left are ICAP-MME mean and spread of total/fine/coarse AODs for the analysis mode. The two columns on the right are the difference between the forecast mode and the analysis mode. Global mean values of the ensemble mean/spread and the differences are also shown for each size mode and forecast mode.
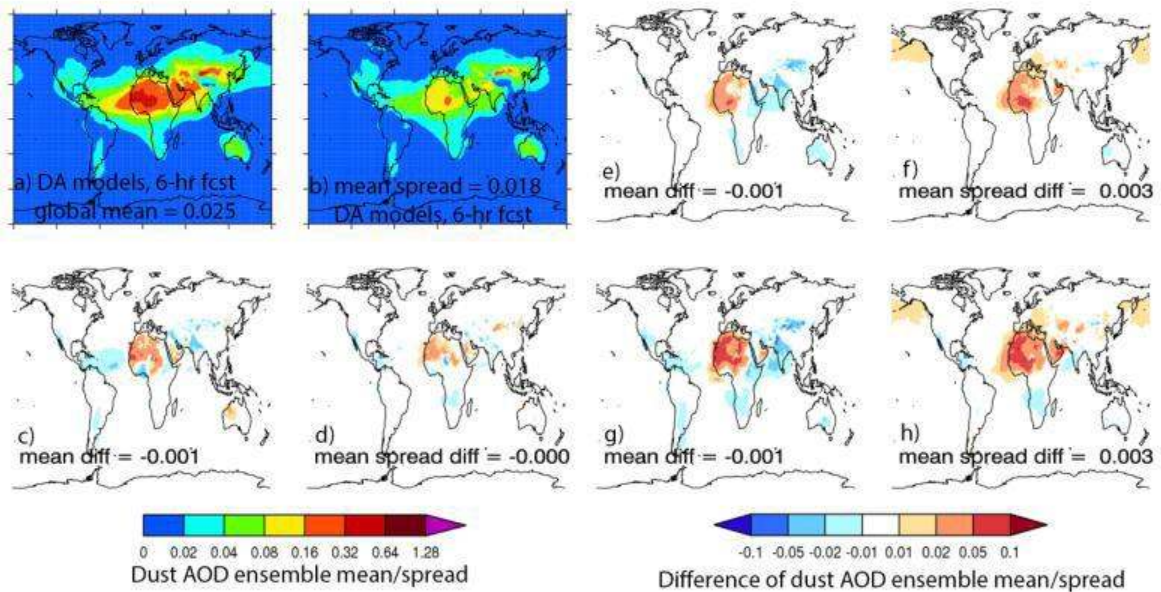
Figure 9. Average ensemble mean and spread of dust AOD at 550 nm of the ICAP DA models at their analysis mode, and the differences from their forecast mode, and from all models at their analysis and forecast modes for Jun 2016-May 2017. a, b) ensemble mean and spread of the 6-hr forecasts of the DA models; c, d) difference from the 72-hr forecast of the DA models; e, f) difference from the 6-hr forecasts of all 7 dust models. g, h) difference from the 72-hr forecasts of all 7 dust models.
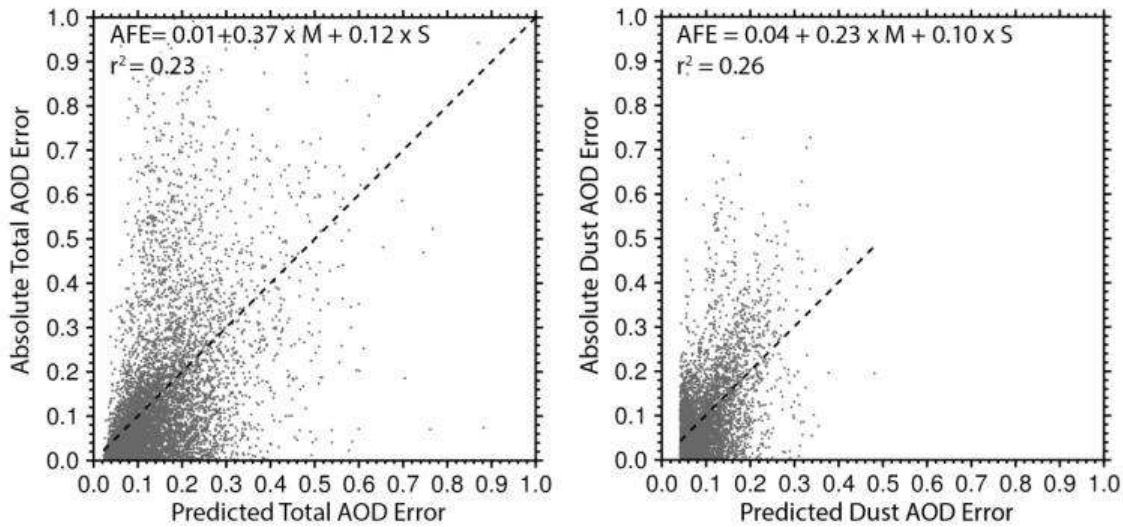
Figure 10. ICAP MME absolute AOD forecast error (AFE) of the 72-hr forecasts versus predicted error using the multivariate regression equations listed in Table 5 and 6. a) Total AOD, b) Dust AOD. "M" stands for ensemble mean and "S" for ensemble spread in the regression relations inside of the plotting area.
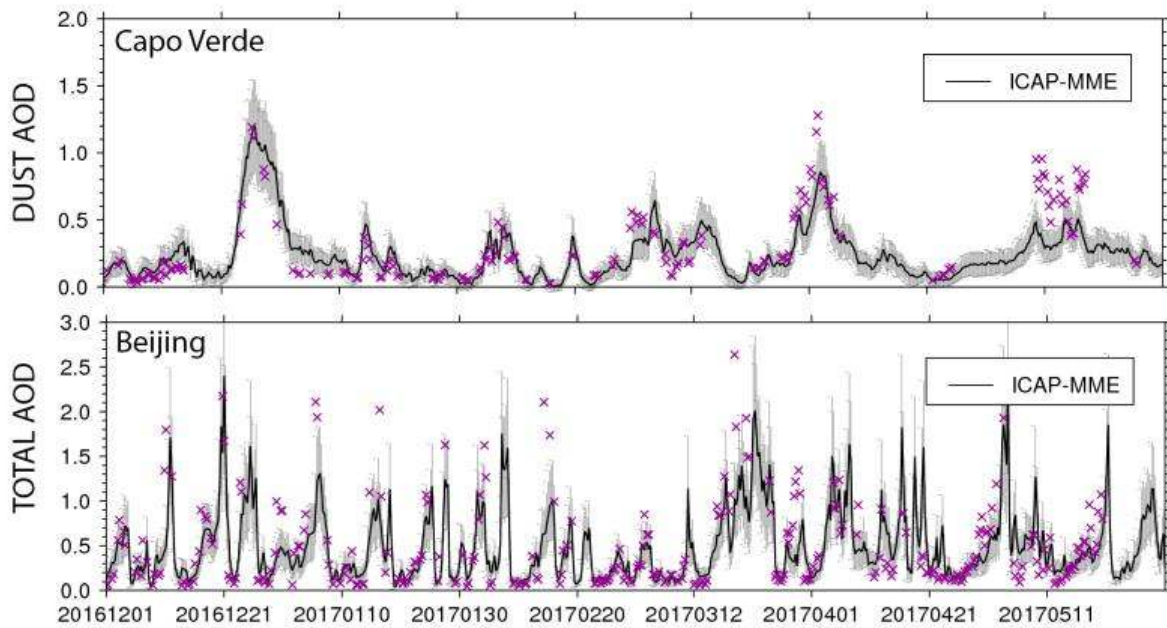
Figure 11. Time series of AERONET AOD and ICAP-MME consensus mean AOD with forecast error estimate derived from the multivariate regression models for the 72-hr forecasts of a) Dust AOD at Capo Verde, b) Total AOD at Beijing. Magenta crosses indicate AERONET AOD observations.

	2012	2013	2014	2015	2016	2017
Yonsei_University	0.47	0.46	0.54	0.42	0.45	0.42
Mezaira	0.54	0.36	0.33	0.39	0.35	0.35
Singapore	0.44	0.42	0.51	0.72	0.29	0.27
Rio_Branco	0.18	0.11	0.16	0.24	0.23	0.27
Ragged_Point	0.17	0.16	0.16	0.19	0.15	0.20
Palma_de_Mallorca	0.17	0.10	0.16	0.13	0.13	0.13
Monterey	0.08	0.07	0.07	0.07	0.28	0.13
Moldova	0.18	0.17	0.17	0.14	0.17	0.16
Minsk	0.16	0.16	0.15	0.14	0.14	0.14
Kanpur	0.62	0.66	0.63	0.62	0.65	0.64
Ilorin	0.76	0.53	0.66	0.67	0.75	0.77
GSFC	0.15	0.12	0.13	0.13	0.10	0.11
Gandhi_College	0.64	0.61	0.58	0.64	0.59	0.64
Chiang_Mai_Met_Sta	0.47	0.62	0.45	0.41	0.60	0.34
Chapais	0.12	0.11	0.09	0.10	0.09	0.05
Cart_Site	0.12	0.11	0.11	0.11	0.08	0.12
Capo_Verde	0.36	0.28	0.30	0.37	0.36	0.44
Beijing	0.70	0.59	0.66	0.61	0.52	0.48
Banizoumbou	0.54	0.41	0.38	0.55	0.56	0.48
Amsterdam_Island	NaN	NaN	0.15	0.11	0.07	0.07
Alta_Floresta	0.23	0.13	0.19	0.28	0.17	0.52

AOD RMSE
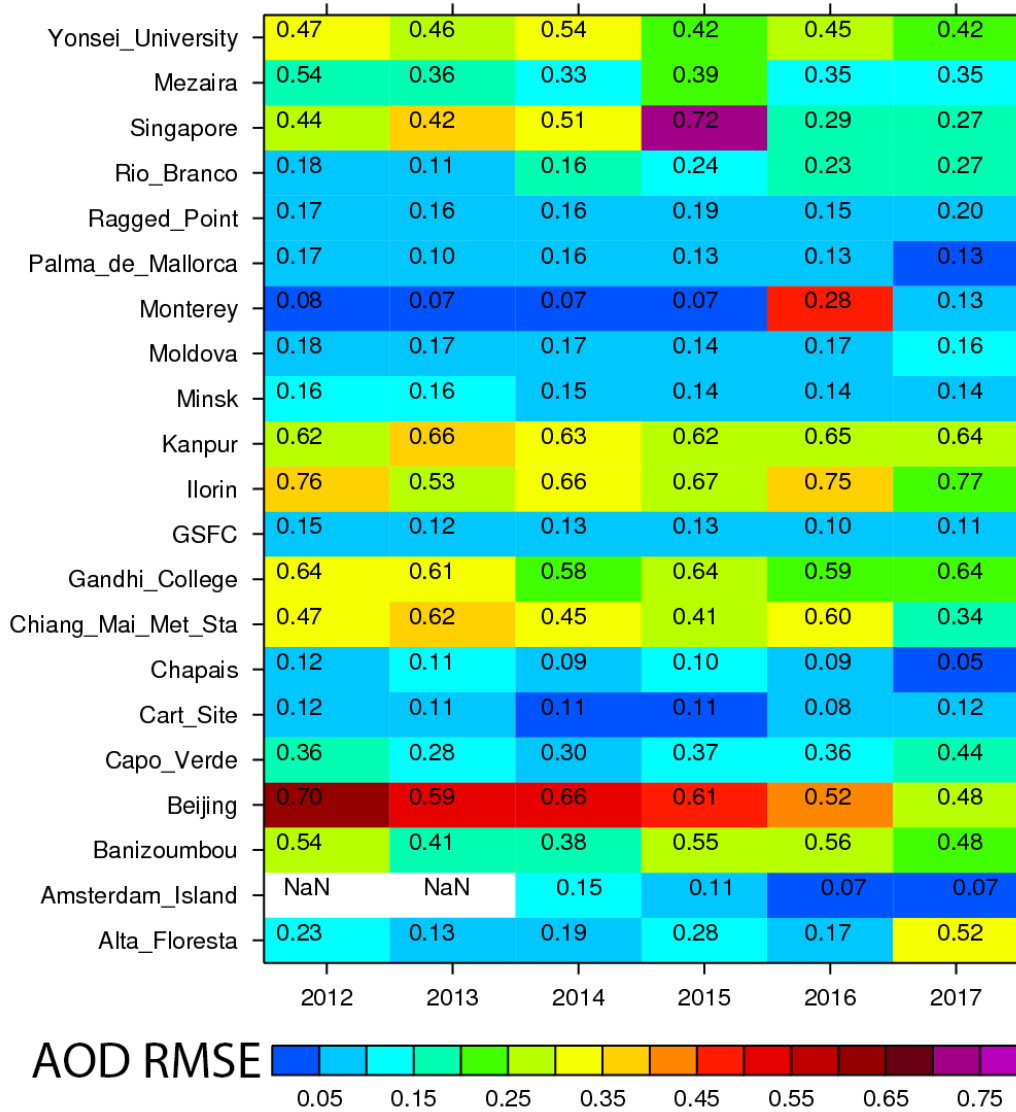
0.05 0.15 0.25 0.35 0.45 0.55 0.65 0.75

Figure 12. Evolution of ICAP-MME performance in terms of RMSE of total AOD at 550 nm of the 6-hr forecasts for the 21 AERONET sites shown in Table 1, over 2012-2017 time period. The number inside of each grid represents yearly mean AERONET V3L1.5 total AOD at 550 nm for the site and year. RMSE is shown in color. "NaN" means not enough data points (at least 100) for evaluation.
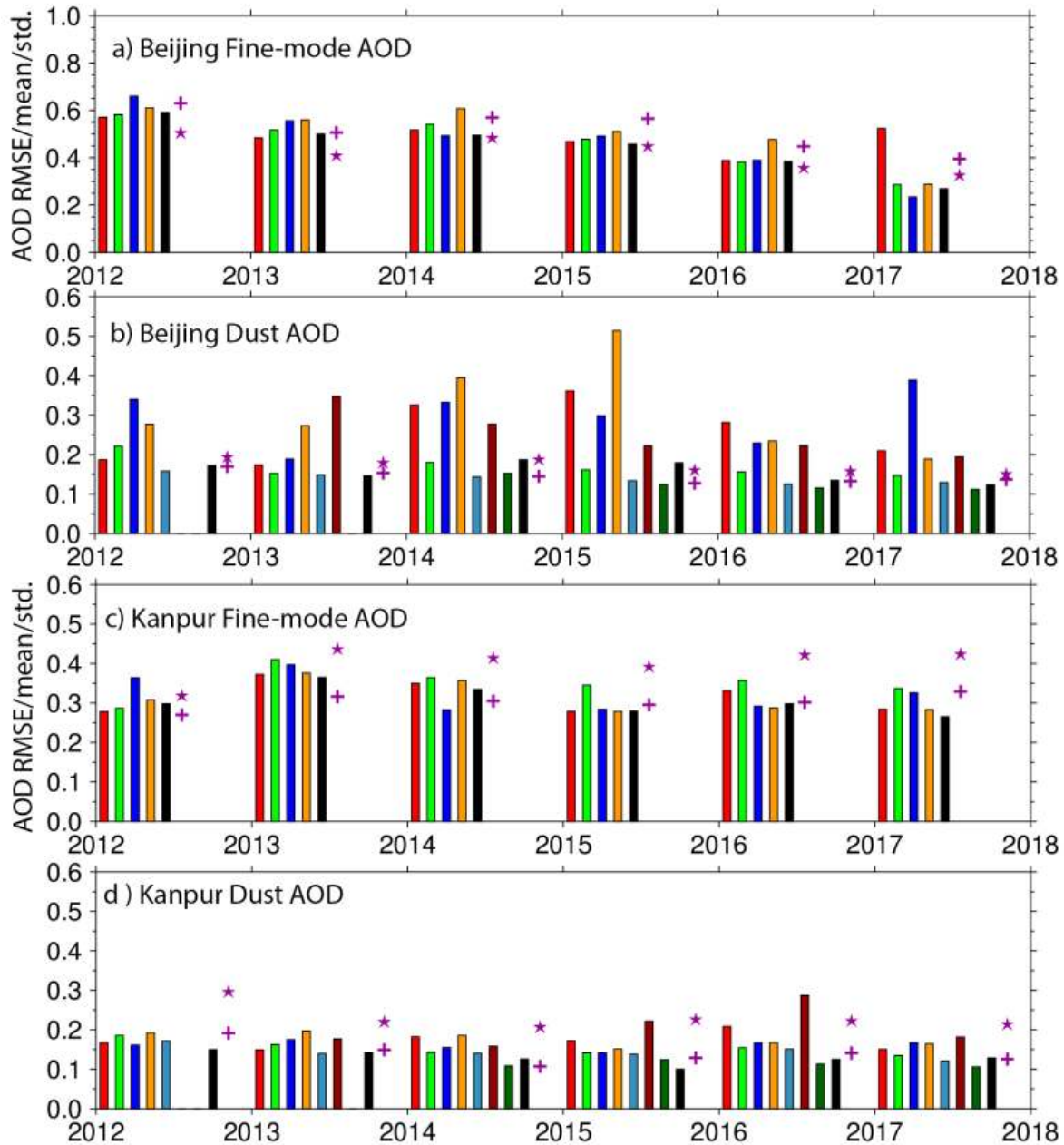
Figure 13. Evolution of ICAP individual models and MME fine-mode and dust 550 nm AOD RMSE of the 72-hr forecasts at Beijing and Kanpur over 2012-2017. Individual models are in color bars and ICAP-MME in black bars. Models 1 to 7 are in red, green, blue, orange, light blue, scarlet and dark green, respectively, which are also in sequence in the dust AOD

histograms. Also shown are AERONET yearly mean fine-mode and coarse-mode AOD in purple
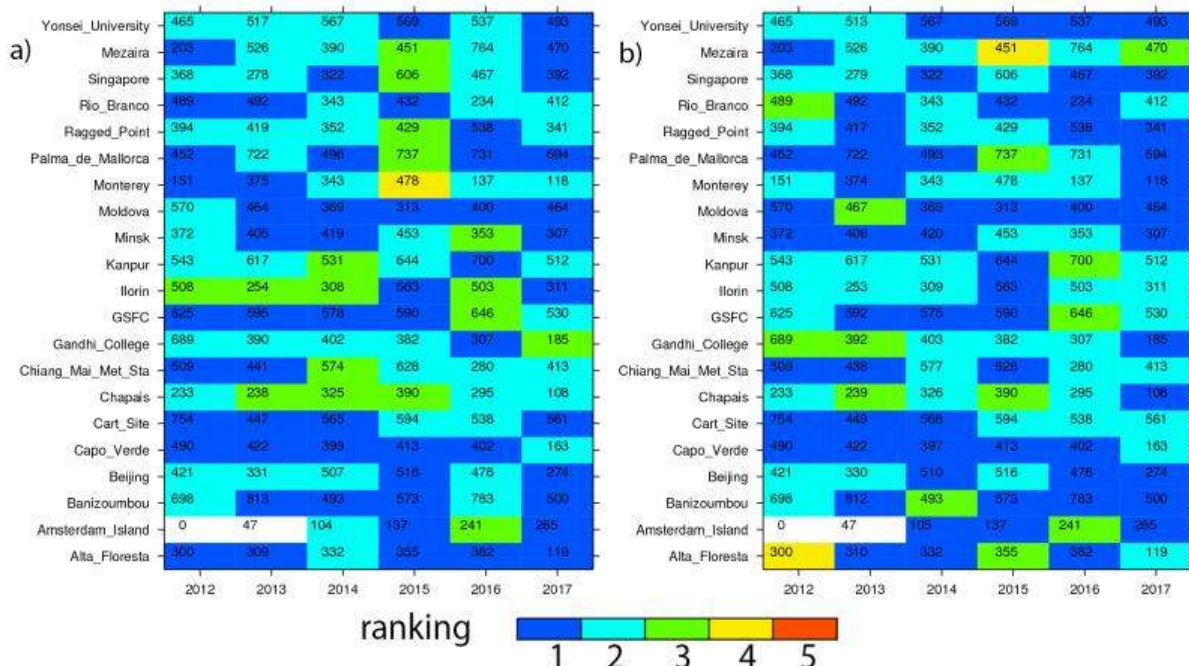
pluses and standard deviation in purple stars.

Figure 14. Ranking of ICAP-MME among all the models in terms of total AOD RMSE over 2012-2017 for a) the 6-hr forecasts and b) the 72-hr forecasts. The number inside of each grid represents total number of paired 6-hrly AERONET and ICAP-MME data for the site and year. Ranking of ICAP-MME is shown in color. This is basically what is shown in Table 2, except for consecutive years.