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Interpretation of geostationary satellite aerosol optical depth (AOD) over East Asia in relation to fine particulate matter ($PM_{2.5}$): insights from the KORUS-AQ aircraft campaign and seasonality

Shixian Zhai[1], Daniel J. Jacob[1], Jared F. Brewer[1], Ke Li[1], Jonathan M. Moch[1], Jhoon Kim[2, 3], Seoyoung Lee[2], Hyunkwang Lim[2], Hyun Chul Lee[3], Su Keun Kuk[3], Rokjin J. Park[4], Jaein I. Jeong[4], Xuan Wang[5], Pengfei Liu[6], Gan Luo[7], Fangqun Yu[7], Jun Meng[8, a], Randall V. Martin[8], Katherine R. Travis[9], Johnathan W. Hair[9], Bruce E. Anderson[9], Jack E. Dibb[10], Jose L. Jimenez[11], Pedro Campuzano-Jost[11], Benjamin A. Nault[11, b], Jung-Hun Woo[12], Younha Kim[13], Qiang Zhang[14], Hong Liao[15]

[1]Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA

[2]Department of Atmospheric Sciences, Yonsei University, Seoul, Republic of Korea

[3]Samsung Particulate Matter Research Institute, Samsung Advanced Institute of Technology, 130 Samsung-ro, Yeongtong-gu, Suwon-si, Gyeonggi-do, Republic of Korea

[4]School of Earth and Environmental Sciences, Seoul National University, Seoul, Republic of Korea

[5]School of Energy and Environment, City University of Hong Kong, Hong Kong SAR, China

[6]School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA, USA

[7]Atmospheric Sciences Research Center, University at Albany, Albany, New York, USA

[8]Department of Energy, Environmental & Chemical Engineering, Washington University in St Louis, MO, USA

[9]NASA Langley Research Center, Hampton, VA, USA

[10]Institute for the Study of Earth, Oceans, and Space, University of New Hampshire, Durham, NH, USA

[11]Department of Chemistry, and Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, USA

[12]Department of Civil and Environmental Engineering, Konkuk University, Seoul, Republic of Korea

[13]International Institute for Applied Systems Analysis (IIASA), 2361 Laxenburg, Austria

[14]Department of Earth System Science, Tsinghua University, Beijing, China.

[15]Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, School of Environmental Science and Engineering, Nanjing University of Information Science and Technology, Nanjing, China.

[a] Now at Department of Atmospheric & Oceanic Sciences, University of California, Los Angeles, California, USA

[b] Now at Center for Aerosol and Cloud Chemistry, Aerodyne Research, Inc., Billerica, MA, USA

Correspondence: Shixian Zhai (zhaisx@g.harvard.edu)

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Abstract. Geostationary satellite sensors over East Asia (GOCI and AHI) are now providing continuous mapping of aerosol optical depth (AOD) at 550 nm to improve monitoring of fine particulate matter ($PM_{2.5}$) air quality. Here we evaluate our understanding of the physical relationships between AOD and $PM_{2.5}$ over East Asia by using the GEOS-Chem atmospheric chemistry model to simulate observations from multiple sources: 1) the joint NASA-NIER Korea - United States Air Quality aircraft campaign over South Korea (KORUS-AQ; May-June 2016); 2) AODs from the AERONET ground-based network; 3) AOD from a new GOCI/AHI fused product; and 4) surface $PM_{2.5}$ networks in South Korea and China. The KORUS-AQ data show that 550 nm AOD is mainly contributed by sulfate-nitrate-ammonium (SNA) and organic aerosols in the planetary boundary layer (PBL), despite large dust concentrations in the free troposphere, reflecting the optically effective size and the high hygroscopicity of the PBL aerosols. Although GEOS-Chem is successful in reproducing the KORUS-AQ vertical profiles of aerosol mass, its ability to link AOD to $PM_{2.5}$ is limited by under-accounting of coarse PM and by a large overestimate of nighttime $PM_{2.5}$ nitrate. A broader analysis of the GOCI/AHI AOD data over East Asia in different seasons shows agreement with AERONET AODs and a spatial distribution consistent with surface $PM_{2.5}$ network data. The AOD observations over North China show a summer maximum and winter minimum, opposite in phase to surface $PM_{2.5}$. This is due to low PBL depths compounded by high residential coal emissions in winter, and high relative humidity (RH) in summer. Seasonality of AOD and $PM_{2.5}$ over South Korea is much weaker, reflecting weaker variation of PBL depth and lack of residential coal emissions. Physical interpretation of the satellite AOD data in terms of surface $PM_{2.5}$ is sensitive to accurate information on aerosol size distributions, PBL depths, RH, the role of coarse particles, and diurnal variation of $PM_{2.5}$.

1 Introduction

$PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm) in surface air is a severe public health concern in East Asia. Surface monitoring networks of $PM_{2.5}$ are too sparse to assess population exposure and satellite observations of aerosol optical depth (AOD) can provide a valuable complement with continuous mapping (Van Donkelaar et al., 2015). The value of satellite observations is now increasing with the advent of geostationary satellite sensors, including the Geostationary Ocean Color Imager (GOCI) launched by the Korea Aerospace Research Institute (KARI) in 2011 (Choi et al., 2016; Lee et al., 2019) and the Advanced Himawari Imager (AHI) launched by the Japan Aerospace Exploration Agency (JAXA) in 2014 (Lim et al., 2018). There is a need to better understand the physical relationships between AOD and $PM_{2.5}$. Here we use the GEOS-Chem chemical transport model (CTM) to analyze and simulate the AOD-$PM_{2.5}$ relationships over East Asia, exploiting vertical profiles from the joint NASA-NIER Korea - United States Air Quality (KORUS-AQ) field study in May-June 2016 (Crawford et al., 2021; Peterson et al., 2019; Jordan et al., 2020) as well as GOCI/AHI geostationary satellite data compared to surface observation networks.

A number of past studies have used satellite AOD data to infer surface $PM_{2.5}$ using physical and statistical models. AOD measures aerosol extinction (scattering and absorption) integrated over the atmospheric column, so that the relationship to $PM_{2.5}$ depends on the vertical distribution and optical properties of aerosols. The standard geophysical

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approach has been to use a CTM, such as GEOS-Chem, to compute the $PM_{2.5}$/AOD ratio (Liu et al., 2004), with recent applications correcting for CTM biases using available $PM_{2.5}$ surface network data (Van Donkelaar et al., 2016; Hammer et al., 2020). An alternative approach is to use machine-learning algorithms to relate satellite AOD to $PM_{2.5}$ by training on the surface network data (Wei et al., 2021), and sometimes including CTM values as predictors (Di et al., 2019). Yet another approach is to assimilate the satellite-measured AODs in a CTM and correct in this manner the $PM_{2.5}$ simulation, although this requires attribution of model AOD errors to specific model parameters (Kumar et al., 2019; Saide et al., 2014; Sekiyama et al., 2010; Cheng et al., 2019). In all of these approaches, a better understanding of the physical relationship of AOD-$PM_{2.5}$ can greatly enhance the capability to infer $PM_{2.5}$ from AOD data.

2 Data and methods

2.1 Observations

We use observations over China and South Korea from multiple platforms including surface sites, aircraft, and satellites (Table 1 and 2). Surface data (Table 1) include $PM_{2.5}$ from national observation networks in China (Zhai et al., 2019) and South Korea (Jordan et al., 2020), speciated $PM_{2.5}$ at 7 supersites in South Korea during KORUS-AQ (Choi et al., 2019), and ground-based AODs from the AERONET network at 2 sites in North China and 10 sites in South Korea (21 sites during KORUS-AQ). We use total and fine-mode AODs at 500 nm wavelength from the AERONET Version 3 Level 2.0 database (Giles et al., 2019). The AERONET AODs at 500 nm are converted to 550 nm using Ångström Exponents at 500 nm for consistency with the satellite AOD data.

Table 1. Surface site observations used in this work (2016)

Variables	Number of sites
$PM_{2.5}$ in North China [a]	117
$PM_{2.5}$ in South Korea [b]	130
$PM_{2.5}$ composition in South Korea (May-June 2016) [c]	7
AERONET total and fine mode AOD in East China [d]	5
AERONET total and fine mode AOD in South Korea [d]	10-21 [e]

[a] Hourly $PM_{2.5}$ from the China National Environmental Monitoring Centre (CNEMC; quotsoft.net/air/) in North China (115.5-122° E, 34.5-40.5° N), including only sites with more than 90% data coverage in each month of 2016. Quality control of the CNEMC dataset is described in our previous study (Zhai et al., 2019). The $PM_{2.5}$ measurements are made at reference $RH \leq 35\%$.

[b] Hourly $PM_{2.5}$ from the AirKorea network (airkorea.or.kr), with the same data selection criteria as for North China. The $PM_{2.5}$ measurements are made at reference $RH \leq 35\%$.

[c] Major $PM_{2.5}$ components including sulfate, nitrate, ammonium, organic carbon, and black carbon at 7 supersites in South Korea during KORUS-AQ (May-June 2016; Choi et al., 2019). The mass concentration of organic carbon is converted to that of organic aerosol with a multiplicative factor of 1.8 based on KORUS-AQ observations (Kim et al., 2018).

[d] AODs are from the AERONET Version 3 Level 2.0 all-points database (aeronet.gsfc.nasa.gov), except that AODs at the XuZhou site in East China are from the Version 3 Level 1.5 database. AOD at 500 nm (AOD_{500nm}) is converted to 550 nm (AOD_{550nm}) using Ångström Exponent at 500 nm (AE_{500nm}) following: $\mathrm{AOD}_{550\mathrm{nm}} = \mathrm{AOD}_{500\mathrm{nm}}\left(\frac{550}{500}\right)^{-\mathrm{AE}_{500\mathrm{nm}}}$.

[e] AERONET AODs in South Korea are from 10 sites for the full year of 2016 and 21 sites during KORUS-AQ.

The KORUS-AQ campaign (Table 2) includes 20 flights over the Korean peninsula and the surrounding ocean from May 2 to June 10, 2016, with vertical profiling up to 8 km altitude. We use the aircraft observations of remote and in situ aerosol extinction (scattering + absorption) coefficients, dry aerosol number size distributions, sub-micron non-refractory aerosol composition, bulk aerosol ionic composition, black carbon (BC), and relative humidity (RH).

Geostationary satellite AOD data are from the GOCI and AHI instruments, covering eastern China and South Korea at 6 km × 6 km spatial resolution and hourly temporal resolution for 8 hours per day (09:30 to 16:30 local time). We use the fused AOD product generated from the GOCI and AHI AOD retrievals, each using two different surface reflectance methods (Lim et al., 2020). Fusion of this four-member ensemble is done by the maximum likelihood estimate (MLE) method, with weighting and averaging based on errors determined by comparison to AERONET AOD. The fused satellite AOD product is shown by Lim et al. (2020) to have higher accuracy than its member products in comparison with AERONET data during the KORUS-AQ campaign. We will refer to it as the 'GEO satellite AOD' product in what follows.

Table 2. KORUS-AQ aircraft observations used in this work (May-June 2016).

Variables	Instruments
Aerosol extinction profile at 532 nm	HSRL[a]
Aerosol scattering coefficient at 550 nm	TSI nephelometers[b]
Aerosol absorption coefficient at 532 nm	PSAPs[c]
Aerosol dry size distribution	TSI LAS[d]
Bulk aerosol ionic composition	SAGA[e]
Sub-micron non-refractory aerosol composition	HR-ToF-AMS[f]
Black carbon concentration	HDSP2[g]
Relative humidity	DLH[h]

[a] NASA Langley airborne High Spectral Resolution Lidar (HSRL) (Hair et al., 2008; Scarino et al., 2014).

[b] NASA Langley TSI-3563 nephelometers (Ziemba et al., 2013).

[c] Radiance Research 3-wavelength particle soot absorption photometers (PSAPs; Ziemba et al., 2013).

[d] Optical size distribution over the 0.1-5.0 µm diameter range from the TSI Laser Aerosol Spectrometer (LAS) Model 3340. Here we follow Nault et al. (2018) and Saide et al. (2020) and multiply the LAS optical equivalent diameters by 1.115 to approximate the geometric diameter.

[e] University of New Hampshire (UNH) Soluble Acidic Gases and Aerosol (SAGA) instrument (Dibb et al., 2003). The cutoff aerodynamic diameter of the inlet is around 4 µm, corresponding to a geometric particle diameter of 2.5 µm (Mcnaughton et al., 2007; Mcnaughton et al., 2009).

[f] University of Colorado Boulder High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS; DeCarlo et al., 2006; Nault et al., 2018; Guo et al., 2020).

[g] NOAA Humidified-Dual-Single-Particle Soot Photometer (HDSP2; Lamb et al., 2018).

[h] NASA Diode Laser Hygrometer (DLH; Podolske et al., 2003).

2.2 GEOS-Chem simulation

We use GEOS-Chem version 12.7.1 (DOI: 10.5281/zenodo.3676008) in a nested-grid simulation at a horizontal resolution of 0.5° × 0.625° over East Asia (100-145 °E, 20-50 °N). GEOS-Chem simulates detailed tropospheric oxidant-aerosol chemistry and is driven here by GEOS-FP assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO). Boundary layer mixing uses the non-local scheme implemented by Lin and McElroy (2010). Dry deposition of gases and particles follows a standard resistance-in-series scheme (Zhang et al., 2001; Fairlie et al., 2007; Fisher et al., 2011; Jaeglé et al., 2018). Wet deposition of gases and particles includes contributions from rainout, washout, and scavenging in convective updrafts (Liu et al., 2001; Amos et al., 2012; Q. Wang et al., 2011; Q. Wang et al., 2014) with recent updates by Luo et al. (2019, 2020). We use pre-archived initial

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conditions from Zhai et al. (2021) and run the model from December 1, 2015 to December 31, 2016. The first month is used for spin-up and the year 2016 is used for analysis.

GEOS-Chem has been used extensively to simulate $PM_{2.5}$ and its composition in East Asia (Geng et al., 2017; Li et al., 2016; Choi et al., 2019; Jeong et al., 2008; Park et al., 2021; Zhai et al., 2021). Here we use the bulk representation of aerosols including sulfate (Park Rokjin et al., 2004; Alexander et al., 2009), nitrate (Jaeglé et al., 2018), primary and secondary organics (Pai et al., 2020), BC (Q. Wang et al., 2014), natural dust in four advected size ranges (Fairlie et al., 2007), anthropogenic fine dust (Philip et al., 2017), and sea salt in two size ranges (Jaeglé et al., 2011). Heterogeneous sulfate formation on aqueous aerosols is represented by a simplified parameterization scheme (Y. Wang et al., 2014), where the SO_2 uptake coefficient (γ) linearly increases from 1×10^{-5} at RH $\leq$ 50% to 2×10^{-5} at RH = 100%. The thermodynamic equilibrium of sulfate-nitrate-ammonium (SNA) aerosols with the gas phase is computed with ISORROPIA II (Fountoukis and Nenes, 2007; Pye et al., 2009) assuming an aqueous aerosol. We include reactive uptake on dust of acid gases (HNO_3, SO_2, and H_2SO_4), limited by consumption of dust alkalinity (Fairlie et al., 2010). The alkalinity of emitted dust is estimated by assuming 7.1% Ca^{2+} and 1.1% Mg^{2+} as alkaline cations by dust mass (Shah et al., 2020).

Monthly anthropogenic emissions are from the Multi-resolution Emission Inventory in 2016 for China (MEIC; Zheng et al., 2018; http://meicmodel.org) and from the KORUSv5 emission inventory at base year 2015 (Woo et al., 2020; http://aisl.konkuk.ac.kr/#/emission_data/korus-aq_emissions) for other Asian countries and shipping emissions. MEIC over China applies weekly and diurnal scaling factors for all anthropogenic emissions (Zheng et al., 2018). The KORUSv5 agricultural NH_3 emissions apply the diurnal scaling factors from MEIC. Natural emissions include NO_x from lightning (Murray et al., 2012) and soil (Hudman et al., 2012), MEGANv2 biogenic volatile organic compounds (VOCs) (Guenther et al., 2012), dust (Meng et al., 2020), and sea salt (Jaeglé et al., 2011). Open fire emissions are from the Global Fire Emissions Database version 4 (GFED4; van der Werf et al., 2017).

2.3 AOD simulation

AOD in GEOS-Chem is diagnosed by integrating vertically the aerosol scattering and absorption coefficients obtained with a standard Mie calculation applied to assumed size distributions, hygroscopicity, refractive indices, and densities for individual aerosol components, and summing over all components (Martin et al., 2003). Optical properties are listed in Table 3. Sulfate, nitrate, and ammonium share the same optical properties and are lumped as an SNA aerosol component for the purpose of optical calculations. All aerosol components except dust are assumed to follow log-normal size distributions. Dust includes 7 size bins (centered at radii of 0.15, 0.25, 0.4, 0.8, 1.5, 2.5, and 4.0 μm) for optical calculations, with the smallest four bins partitioned by mass from the first advected dust bin (< 2.5 μm in geometric diameter) following L. Zhang et al. (2013). Dust particles follow a gamma size distribution within their optical size bins (Curci, 2012). The BC absorption enhancement from coating is as given by X. Wang et al. (2014).

Our initial simulations indicated that aerosol extinction coefficients from the standard GEOS-Chem version 12.7.1 underestimated in situ measured extinction coefficients during KORUS-AQ by 20% on average (Figure S1). We traced this problem to bias in the assumed size distributions for SNA and organic aerosol, as shown in Section 3 (and in Supplementary text and Figure S1-S3). Therefore, we re-computed the diagnostic AOD using updated log-normal size distributions for SNA and organic aerosol with number median radius $R_{N,med} = 0.11$ µm and geometric standard deviation $\sigma = 1.4$, as compared to $R_{N,med} = 0.058$ µm and $\sigma = 1.6$ in the standard model version 12.7.1.

Table 3. Aerosol optical properties [a].

Aerosol component	$R_{N,med}$, µm	σ	Hygroscopicity [b]	Refractive index	ρ, g cm^{-3}
SNA [c]	0.11	1.4	$\kappa = 0.61$	$1.53 - 6.0 \times 10^{-3}i$	1.7
Organic [c]	0.11	1.4	$\kappa = 0.1$	$1.53 - 6.0 \times 10^{-3}i$	1.3
BC	0.020	1.6	GADS	$1.75 - 4.4 \times 10^{-3}i$	1.8
Sea salt (fine)	0.085	1.5	GADS	$1.5 - 1.0 \times 10^{-3}i$	2.2
Sea salt (coarse)	0.40	1.8	GADS	$1.5 - 1.0 \times 10^{-3}i$	2.2
Dust	7 size bins	NA	$\kappa = 0$ [d]	$1.558 - 1.4 \times 10^{-3}i$	2.5-2.65 [e]

[a] Aerosol optical properties used in this work for computing aerosol scattering and absorption coefficients. Values are from the standard GEOS-Chem model version 12.7.1, except for the size distributions of SNA and organic aerosol (see text). All aerosol components except dust have log-normal dry size distributions where $R_{N,med}$ is the number median radius and σ is the geometric standard deviation. Refractive indices are for 550 nm wavelength. ρ is the dry aerosol mass density.

[b] Hygroscopic growth for SNA and organic aerosol as a function of relative humidity (RH, %) is computed from κ-Kohler theory as a diameter growth factor $GF = (1 + \kappa * RH/(100-RH))^{1/3}$ (Latimer and Martin, 2019). Hygroscopic growth factors for other aerosol components are from the Global Aerosol Data Set (GADS) as tabulated in Chin et al. (2002) and Martin et al. (2003).

[c] $R_{N,med}$ and σ are fit to KORUS-AQ observations as described in the text. Standard GEOS-Chem v12.7.1 assumes $R_{N,med} = 0.058$ µm, $\sigma = 1.6$ (Latimer and Martin, 2019).

[d] Hygroscopic growth of dust particles is assumed negligible.

[e] Sub-micron dust particles have a density of 2.5 g cm^{-3} while coarse mode dust particles have a density of 2.65 g cm^{-3}. Dust size distribution is described in the text.

3 Aerosol concentrations and optical properties during KORUS-AQ

Here we use the KORUS-AQ aircraft observations and their simulation with GEOS-Chem to better understand the vertical distributions of different aerosol components contributing to AOD over South Korea. We begin with the mean vertical profile of aerosol mass and go on to examine the aerosol optical properties. This provides the basis for analyzing the observed vertical profile of aerosol extinction, its simulation by GEOS-Chem, and the consistency with GEO satellite and AERONET AOD measurements over South Korea during the KORUS-AQ period.

3.1 Vertical profile of aerosol mass

Figure 1 shows the mean vertical profiles of aerosol mass observed during KORUS-AQ and their simulation by GEOS-Chem. Here and elsewhere, the model is sampled along the flight tracks and at the flight times. The observed vertical distribution of aerosol mass concentrations (Figure 1a) shows that 58% of column aerosol mass is below 2 km altitude, which we define as the planetary boundary layer (PBL), and 34% is at 2-5 km altitude, which we define as the lower free troposphere (FT). The model has a similar vertical distribution (Figure 1b), with 57% of aerosol mass in the PBL and 36% in the lower FT. SNA, organic, and dust each contribute about a third of aerosol mass in the PBL while dust dominates in the lower FT both in the observations and in the model. The enhanced dust in the lower FT is driven by a few dust events, which the model reproduces (Figure S2). Black carbon and sea salt (not shown) make only minor contributions to aerosol mass. The model underestimates sulfate by 28% in the PBL, which leads to a 20% overestimate of nitrate, with canceling effect on the SNA mass simulation.

The GEOS-Chem simulation of organic aerosol in this work uses the simple scheme of Pai et al. (2020) and underestimates aircraft observations by 16% in the PBL. Over 90% of GEOS-Chem organic aerosol is secondary, consistent with observations (Figure S4; Nault et al., 2018; Pai et al., 2020). GEOS-Chem simulation of the KORUS-AQ aerosol component profiles for different meteorological regimes is presented in Park et al. (2021).

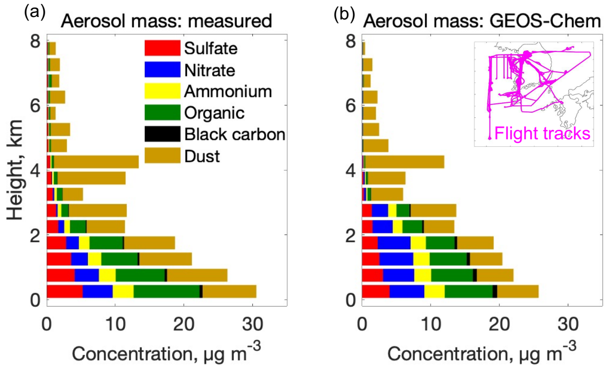

Figure 1. Vertical profiles of aerosol mass during KORUS-AQ. Panel (a) shows the mean vertical distributions of observed mass concentrations of major aerosol components at ambient temperature and pressure. Panel (b) is the same as (a) but from the GEOS-Chem model sampled along the flight tracks (inset). We derive dust concentration from SAGA Ca^{2+} and Na^+ following Shah et al. (2020) by assuming that non-sea salt Ca^{2+} accounts for 7.1% of dust mass: $[dust] = ([Ca^{2+}] - 0.0439\ [Na^+]/2) / 0.071$ where the brackets denote mass concentration. Modeled dust is shown for particles with geometric diameter < 2.5 µm, to be consistent with SAGA measurements (Table 2 footnote e). Measured sulfate, nitrate, ammonium, and organic aerosol concentrations are from the AMS instrument (values from the SAGA instrument are shown in Figure S4). All data are averaged over 500-m vertical bins. Here and elsewhere, we excluded pollution plumes diagnosed by either NO_2 or SO_2 > 10 ppbv (3.4% of all the data).

3.2 Aerosol size distributions

Figure 2a shows the normalized dry aerosol number size distributions on each of the 20 flights and in 3 altitude bands: < 1.5 km, 3-5 km, and 6-7 km (60 lines). The spread in the size distributions above 1 µm in diameter reflects dust influence. We select measurements below 1.5 km altitude when SNA + organic aerosol mass concentrations are more than 4 times that of dust as defining the SNA + organic aerosol size distributions (green lines in Figure 2a). Conditions dominated by SNA + organic aerosols define the lower envelopes of the ensemble of size distributions at diameter > 1 µm. SNA and organics were observed to have similar size distributions during KORUS-AQ (Kim et al., 2018).

Figure 2b converts the SNA + organic dominated number size distributions to volume size distributions. The observed SNA + organic dominated aerosol size distribution is shifted toward larger sizes relative to the standard GEOS-Chem. The secondary maximum in the coarse mode could be due to dust. We fitted the observed SNA + organic aerosol size distributions to a lognormal distribution with volume median radius $R_{V,med}$ = 0.16 µm and geometric standard deviation σ = 1.4. The number median radius is derived from the volume median radius following Seinfeld and Pandis (2016):

$$\ln R_{N,med} = \ln R_{V,med} - 3\ln^2\sigma \qquad (1)$$

which yields $R_{N,med}$ = 0.11 µm. In comparison, the standard GEOS-Chem size distribution from Latimer and Martin (2019) has $R_{N,med}$ = 0.058 µm and σ = 1.6. We adopt the observed log-normal size distribution parameters in what follows (Table 3).

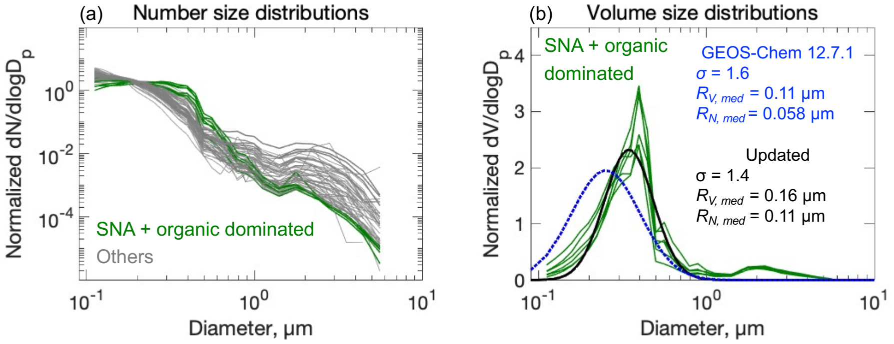

Figure 2. Aerosol dry size distributions measured in the KORUS-AQ aircraft campaign. Panel (a) shows mean normalized number size distributions measured on each of the 20 flights and for 3 altitude bins: < 1.5 km, 3-5 km, and 6-7 km (60 lines total). The SNA + organic dominated size distribution profiles are highlighted in color. Panel (b) shows normalized volume size distributions for conditions dominated by SNA + organic aerosols (green lines), along with a least-square fit to a lognormal distribution (black line), and the standard GEOS-Chem v12.7.1 size distribution from Latimer and Martin (2019) (blue dashed line). Normalization imposes an arbitrary value of unit area below each line. Lognormal distribution parameters are inset in panel (b) including volume median radius ($R_{V,med}$), number median radius ($R_{N,\,med}$), and geometric standard deviation (σ).

3.3 Aerosol extinction and relation to AOD

Figure 3 shows the vertical profiles of ambient aerosol extinction coefficients and RH during KORUS-AQ. Vertical profiles of aerosol extinction were measured on the aircraft both remotely with the HSRL instrument (above and below the aircraft) and in situ with TSI-3563 nephelometers (for scattering) and PSAPs (for absorption). The two agree well, as shown in Figure 3a. They indicate that 76-90% of column aerosol extinction is in the PBL at 0-2 km altitude and 9-19% is in the lower FT at 2-5 km. Both measurements show that aerosol extinction is much more strongly weighted to the PBL than aerosol mass (Figure 1).

Also shown in Figure 3a are the contributions of individual aerosol components to the extinction profile, as computed from the GEOS-Chem optical properties (Table 3) applied to the observed mass concentrations. The sum shows a good match to the measured extinction coefficient profiles. The much larger contribution of the PBL to column aerosol extinction than to column mass is because aerosol mass in the lower FT is mainly composed of dust, whose mass extinction efficiency is much smaller than SNA and organics due to its coarse size and lack of hygroscopic growth (Figure S5). The mean AOD inferred from the aircraft data is 0.36 and is contributed 59% by SNA, 27% by organic aerosol, 12% by dust, and 2% by BC. It is consistent with the mean AODs measured at AERONET stations in South Korea during KORUS-AQ (Figure S6).

Figure 3b shows the GEOS-Chem simulation of aerosol extinction profiles for comparison to the observations in Figure 3a. The model underestimates extinction coefficients by 20% below 1 km altitude, although there is no such underestimate in aerosol mass. This is caused by a negative RH bias in the GEOS-FP meteorological data used to drive GEOS-Chem, particularly at high RH conditions (Figure 3c) and is corrected if we apply the observed RH rather than the GEOS-FP RH to the GEOS-Chem aerosol mass concentrations (Figure 3d).

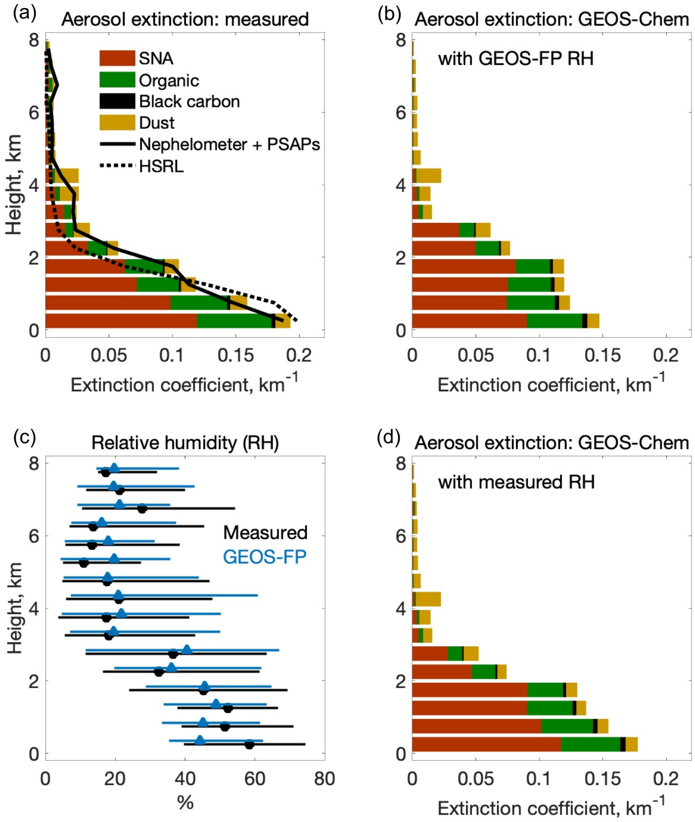

Figure 3. Vertical profiles of aerosol extinction coefficients and relative humidity (RH) during KORUS-AQ. Panel (a) shows the mean observed vertical distributions of 550 nm extinction coefficients measured in situ (nephelometer + PSAPs; at ambient RH) and remotely (HSRL), along with an independent calculation (colored horizontal bars) from the measured mass concentrations of major aerosol components, measured RH, and GEOS-Chem optical properties as given in Table 3. Panel (b) shows the mean aerosol extinction profile in GEOS-Chem and the contributions from the different model components. Panel (c) is the median vertical profile of RH (horizontal bars are 25-75th percentiles) from aircraft measurements and the GEOS-FP assimilated meteorological data used to drive GEOS-Chem. Panel (d) is the same as (b) but calculated using measured RH.

4 AOD and surface particulate matter over South Korea during KORUS-AQ

Our analysis of Section 3 used the KORUS-AQ aircraft data to attribute AOD over South Korea to individual aerosol components and altitudes. We now take the next step of relating satellite to AERONET AODs over the Korea peninsula during KORUS-AQ and evaluating the capability of GEOS-Chem to simulate observed AODs and surface particulate matter concentrations.

Figure 4a shows the spatial distribution of the fused geostationary satellite (GOCI/AHI) AOD (GEO satellite AOD) during the KORUS-AQ period with AERONET AOD added as circles. The GEO satellite AOD shows high values (0.5-0.6) along the west coast of South Korea, significantly correlated with AERONET total AOD with a spatial correlation coefficient (*r*) of 0.7. GEO satellite AOD is biased low at sites in the Seoul Metropolitan Area (SMA) and is biased high on the Yellow Sea islands, resulting in an overall -10% bias, consistent with the validation by Lim et al. (2020). Sampling the AODs at or near the seven $PM_{2.5}$ supersites operating during KORUS-AQ shows no significant bias (inset values in Figure 4a).

Figure 4b-e shows the spatial distributions of GEOS-Chem AOD, surface PM_{10} (particulate matter with aerodynamic diameter less than 10 µm), surface $PM_{2.5}$, and surface coarse PM (PM_{10} minus $PM_{2.5}$; particulate matter with aerodynamic diameter less than 10 µm and larger than 2.5 µm), with surface observations shown as circles and median values at the measurement sites inset. GEOS-Chem reproduces the satellite AOD enhancements along the west coast of South Korea but the values are lower than observed. Comparison of AERONET total and fine mode AOD shows a 13% contribution of coarse particles to total AOD. Comparison of GEOS-Chem to the fine-mode AERONET AOD, as shown in Figure 4b, finds a 15% underestimate that could be attributed to the low-RH bias (Figure 3c). Concurrent measurements of PM_{10} and $PM_{2.5}$ at AirKorea sites show that coarse PM (median 21 $\mu g\ m^{-3}$) accounts for 41% of total PM_{10} (50 $\mu g\ m^{-3}$), while coarse PM in GEOS-Chem is much lower (1.4 $\mu g\ m^{-3}$; Figure 4e). Therefore, the GEOS-Chem underestimate of AOD can mostly be attributed to missing coarse PM. Coarse PM has a concentration larger than 10 $\mu g\ m^{-3}$ across South Korea, with higher concentration in the SMA (~ 30 $\mu g\ m^{-3}$) than in rural areas (~ 15 $\mu g\ m^{-3}$), implying an origin from both anthropogenic and natural sources (Figure 4e).

GEOS-Chem overestimates surface $PM_{2.5}$ by 43% over South Korea (Figure 4d), in contrast to the simulation of AERONET fine mode AOD (Figure 4b). Figure 4f-j shows the spatial distributions of major $PM_{2.5}$ components in GEOS-Chem (background) and measurements (filled circles). GEOS-Chem is not significantly biased relative to the observations for organic aerosol and BC, and underestimates sulfate by 22%. We find that the model bias for $PM_{2.5}$ is largely driven by nitrate, which is overestimated by a factor of 3 and leads to an 56% overestimate of ammonium. By contrast, comparison to the KORUS-AQ data below 1-km altitude showed only a 20% overestimate of nitrate (Figure 1). This is because the model bias is mainly driven by nighttime conditions, as shown in Figure 5. The cause of this large model bias is analyzed by K. R. Travis et al. (manuscript in preparation) and attributed to nighttime nitrate chemistry in the stratified boundary layer.

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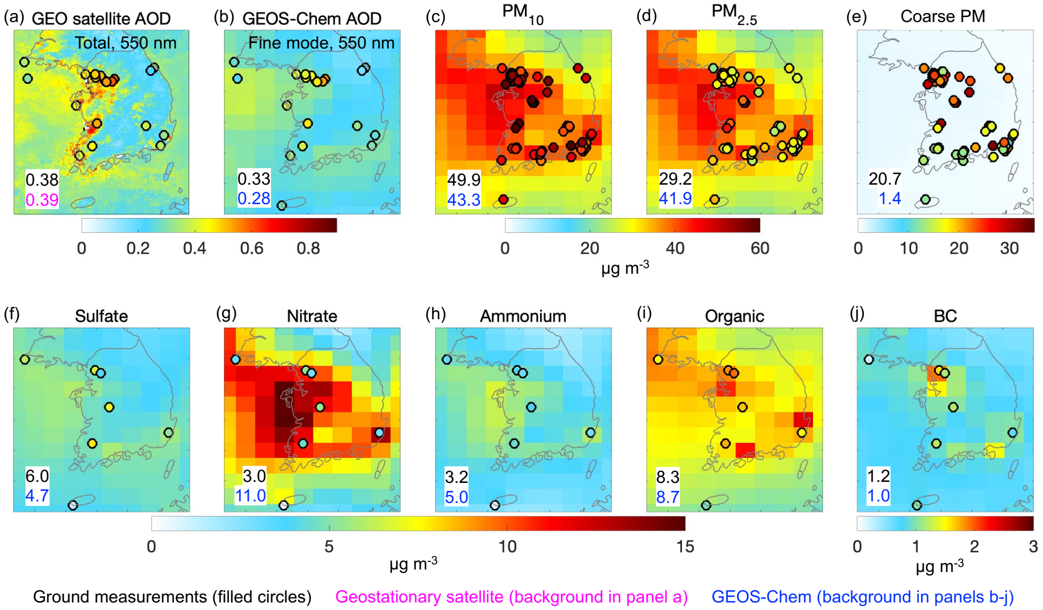

Figure 4. Spatial distributions of AOD, surface PM_{10}, $PM_{2.5}$, coarse PM (PM_{10} minus $PM_{2.5}$), and major $PM_{2.5}$ components over South Korea averaged during KORUS-AQ (May 9 - June 10, 2016). Panel (a) shows the fused geostationary (GEO) 550 nm AOD from the GOCI and AHI satellites (background) and AERONET 550 nm total AOD (filled circles). Panel (b) shows GEOS-Chem 550 nm AOD sampled at hourly GEO satellite AOD (GEOS-Chem clear-sky AOD; background) and AERONET 550 nm fine mode AOD (filled circles). Panel (c) shows surface PM_{10} modelled by GEOS-Chem (background) and measured at ground sites (filled circles). Panels (d-j) are the same as panel (c) but respectively for $PM_{2.5}$, coarse PM (PM_{10} minus $PM_{2.5}$), and sulfate, nitrate, ammonium, organic, and BC $PM_{2.5}$ components. Values inset are median values from ground-based measurements (black) and sampled from GEO satellite (magenta) and GEOS-Chem (blue). Measured PM_{10}, $PM_{2.5}$, and coarse PM in panels (c-e) are shown for a random selection of 50% of AirKorea sites to visualize spatial distribution, and inset values are for the seven supersites where $PM_{2.5}$ composition was measured. Median AOD values inset are sampled at or near the seven supersites to avoid biasing by the large number of sites in the Seoul Metropolitan Area. Modelled total $PM_{2.5}$ concentrations are calculated at 35% RH (Table 3). Modelled PM_{10} is the sum of $PM_{2.5}$, coarse dust, and coarse sea salt.

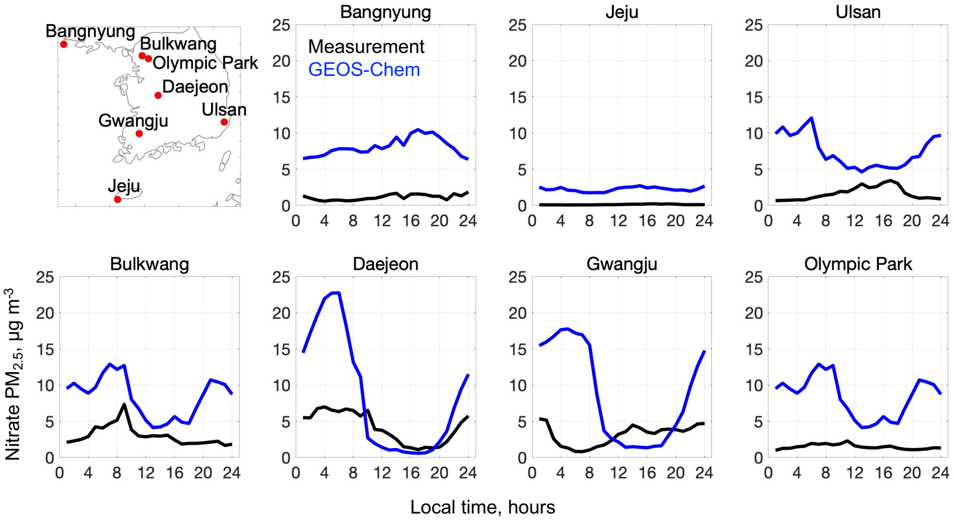

Figure 5. Median diurnal variations of $PM_{2.5}$ nitrate concentrations at the seven supersites (top left panel) operated in South Korea during KORUS-AQ (May 9 - June 10, 2016). Values are medians binned by hour. GEOS-Chem model values are sampled to coincide with the measurements.

5 AOD and its relationship to $PM_{2.5}$ over East Asia

We build on our analysis of the KORUS-AQ period for a broader interpretation of the distribution of AOD over Korea and China and its relationship to surface $PM_{2.5}$, acknowledging that the conditions sampled in KORUS-AQ may not be representative of other seasons or of China. Figure 6 shows the spatial distributions of 2016 annual and seasonal mean geostationary (GEO) satellite AODs, the corresponding GEOS-Chem clear-sky AODs, and GEOS-Chem surface $PM_{2.5}$. The Figure gives normalized mean biases (*NMB*s) relative to ground-based measurements from AERONET and from the $PM_{2.5}$ surface networks (shown as circles).

On an annual mean basis, AOD over North China (~ 0.5-0.6) is about 50% larger than over South Korea (~ 0.3-0.4). AOD over South Korea shows higher values (> 0.4) in the Seoul Metropolitan Area, consistent with that during the KORUS-AQ period (Figure 4a). Transport from the Asian continent is strongest in spring when the frequency of cold front passages is highest (Liu et al., 2003). AERONET total AOD in spring (0.4-0.6) is twice as large as fine-mode AOD (0.2-0.3), reflecting a large contribution of dust. In seasons other than spring, 80-90% of AERONET total AOD is contributed by the fine mode. There is large seasonality in AODs over North China, and weaker seasonality over South Korea, which will be discussed below.

The GEOS-Chem clear-sky AODs show the same spatial and seasonal patterns as GEO satellite AODs but tend to be low in spring and summer. Comparison of the model to AERONET AODs confirms this bias and shows better agreement with fine-mode AOD in spring (*NMB* of -1%), implying an underestimate of coarse dust that is consistent with our comparisons to the AirKorea PM_{10} network data (Figure 4e). Comparison of clear-sky and all-sky AODs in

GEOS-Chem shows no significant difference on an annual and seasonal mean basis, except for winter (Figure S7). Winter has larger all-sky AOD than clear-sky AOD and the lowest rate of successful satellite retrievals (Figure S7), which may be due in part to misclassification of heavy wintertime $PM_{2.5}$ pollution as clouds (Zhang et al., 2020).

The spatial distributions of $PM_{2.5}$ in GEOS-Chem in different seasons match closely the observations (Figure 6, bottom row). We see also a close coincidence between the spatial distributions of $PM_{2.5}$ and AODs, both in the observations and the model. On an annual mean basis, GEOS-Chem overestimates $PM_{2.5}$ by 16% in North China and by 14% in South Korea, even though it underestimates AERONET fine mode AODs by 15%. The overestimate of $PM_{2.5}$ in South Korea is worst in spring (27%), consistent with KORUS-AQ results which we previously attributed to excessive nighttime nitrate build-up in the model. Over North China, the overestimate of $PM_{2.5}$ is worst in summer (33%), consistent with the nitrate overestimate in summer shown in our previous study (Zhai et al., 2021), which could also be due to model overestimate of nighttime nitrate (Miao et al., 2020).

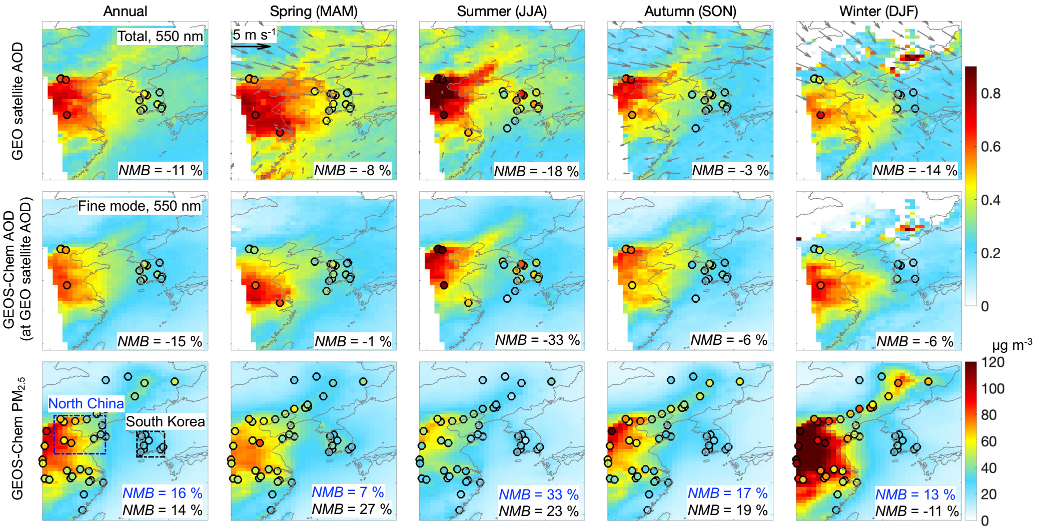

Figure 6. Spatial distributions of 2016 annual and seasonal mean AOD (550 nm) and surface $PM_{2.5}$. The top row shows the observed GOCI/AHI geostationary satellite AOD (GEO satellite AOD) on the GEOS-Chem 0.5° × 0.625° grids with 925 hPa GEOS-FP wind fields and AERONET total AODs (circles). The middle row shows GEOS-Chem AOD sampled in the same way as hourly GEO satellite AOD (GEOS-Chem clear-sky AOD), with AERONET fine mode AOD added as circles. The bottom row shows GEOS-Chem surface $PM_{2.5}$ (background) with surface network measurements (circles). AERONET AODs are shown only when more than 10 months of data are available for the annual mean and all 3 months data are available for each season. The $PM_{2.5}$ observations shown are for a random selection of 7% of network sites for visual clarity. GEOS-Chem $PM_{2.5}$ is calculated at 35% RH (Table 3). Normalized mean biases (*NMB*s) inset are for the comparisons of GEO satellite and GEOS-Chem values to the corresponding ground measurements.

Figure 7 compares the seasonalities of AOD and $PM_{2.5}$ over the North China and South Korea regions. The GEO satellite AOD over North China peaks in July and is minimum in winter. Most of AOD is attributed by GEOS-Chem to SNA aerosol, same as in South Korea. AOD over South Korea also has a summer maximum and winter minimum but with weaker amplitude than over North China. The model is biased low in the summer and this is largely due a low RH bias (Figure S8), as seen previously in the KORUS-AQ comparisons but amplified by the high RH in summer that drives hygroscopic growth (Latimer and Martin, 2019).

Surface $PM_{2.5}$ in the observations over North China and South Korea shows opposite seasonality to AOD, with minimum values in summer and maximum values in winter-spring. GEOS-Chem reproduces the strong seasonality of $PM_{2.5}$ in North China and the much weaker seasonality in South Korea. The high $PM_{2.5}$ values over North China in winter in the model are mostly driven by organic aerosol, reflecting the large residential coal burning source (Figure S9; Zheng et al., 2018). In South Korea, by contrast, household energy is mainly from natural gas and electricity (Lee et al., 2020; Woo et al., 2020). PBL height also shows a stronger seasonality over North China than over South Korea (Figure S8). The mean $PM_{2.5}$/AOD ratio over North China in winter (236 µg m^{-3}) is 8 times that in summer (29 µg m^{-3}), with autumn (94 µg m^{-3}) and spring (89 µg m^{-3}) in between, while over South Korea, the $PM_{2.5}$/AOD ratio in winter (62 µg m^{-3}) is only 70% larger than in summer (36 µg m^{-3}).

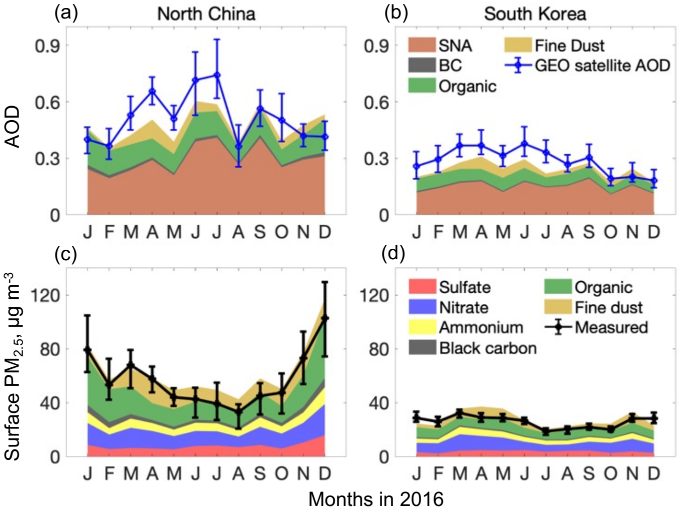

Figure 7. Seasonality of AOD and $PM_{2.5}$ over North China and South Korea, and contributions from individual aerosol components. Lines show regional medians (error bars: 25th and 75th percentiles) for the ensemble of monthly averaged observations in the regions (Figure 6) in 2016. GEOS-Chem values are shown as stacked contours for individual components and are sampled in the same way as the observations.

6 Conclusions

Geostationary satellite observations of aerosol optical depth (AOD) from the GOCI and AHI satellite instruments have tremendous potential for monitoring of $PM_{2.5}$ air quality over East Asia if they can be properly interpreted. Here we used a new fused GOCI/AHI satellite product together with AERONET ground-based AOD measurements, vertical profiles over South Korea from the KORUS-AQ aircraft campaign (May-June 2016), and surface network

PM observations, simulated collectively with the GEOS-Chem transport model, to better understand the physical relationship between satellite AOD and $PM_{2.5}$.

The KORUS-AQ observations show that total aerosol extinction (550 nm) in the vertical column is dominated by sulfate-nitrate-ammonium (SNA) and organic aerosol in the planetary boundary layer (PBL), despite large concentrations of dust in the free troposphere. This reflects the 550 nm optically effective size and high hygroscopicity of the PBL aerosols. Adjustment of GEOS-Chem aerosol optical properties to the observed SNA and organic aerosol size distributions enabled a successful simulation of the aerosol extinction profile, although the simulation is highly sensitive to bias in the relative humidity (RH) of the driving meteorological data. SNA aerosol contributed 59% of column aerosol extinction in the KORUS-AQ data, while organic aerosol contributed 27% and dust contributed 12%.

Comparison of GOCI/AHI geostationary (GEO) satellite AOD to AERONET AODs over South Korea shows good agreement, with high values along the west coast. GEOS-Chem is more consistent with the fine-mode AERONET AOD because of its insufficient accounting of coarse particles, which account for 13% of AERONET AOD. GEOS-Chem overestimates 24-h surface $PM_{2.5}$ over South Korea by 43% during the KORUS-AQ period, despite its successful simulation of the aircraft data and fine-mode AERONET AOD, and we find that this is due to a large overestimate of nighttime nitrate.

Broader examination of the GOCI/AHI AOD satellite data over East Asia shows spatial distributions and magnitudes consistent with AERONET and featuring in particular strong Asian outflow in spring that includes a large dust component. We find that AODs and $PM_{2.5}$ have similar large-scale spatial distributions but opposite seasonality. $PM_{2.5}$ in North China has a strong winter maximum and summer minimum, while the AOD shows the opposite. We find that this is mainly driven by residential coal heating sources and low PBL depths in winter, and high RH in summer. Observations of $PM_{2.5}$ and AOD in South Korea show the same seasonal phases as in North China but with much weaker amplitude, reflecting the lack of residential coal burning in winter and a weaker seasonal amplitude of PBL depth.

In summary, we find that the geostationary GOCI/AHI satellite AOD data provide high-quality information for monitoring of $PM_{2.5}$ over East Asia but that physical interpretation requires accurate information on aerosol size distributions, PBL depths, RH, the role of coarse particles, and diurnal variation of $PM_{2.5}$. Addressing these uncertainties should be a target of future work.

Data availability. Aircraft data during KORUS-AQ are available at: www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq. $PM_{2.5}$ data over China are from: quotsoft.net/air/. $PM_{2.5}$ data over South Korea are from: www.airkorea.or.kr/web. AERONET data can be found at: aeronet.gsfc.nasa.gov. The MEIC emission inventory are at: www.meicmodel.org/. The KORUSv5 emission inventory is developed by Konkuk University, available at: http://aisl.konkuk.ac.kr/#/emission_data/korus-aq_emissions.

Author contributions. SZ and DJJ designed the study. SZ performed the data analysis and model simulations with contributions from JFB, KL, HCL, SKK, XW, PL, KRT, and Hong Liao. JK, SL, and Hyunkwang Lim provided satellite AOD data. RJP and JIJ contributed to AirKorea data processing. JM and RM provided the dust emission inventory. GL, FY, and JMM updated wet deposition simulation. JWH, BEA, JED, JLJ, PCJ, and BAN contributed to KORUS-AQ campaign measurements. JHW and YK provided the KORUSv5 emission inventory. QZ provided the MEIC emission inventory. SZ and DJJ wrote the paper with input from all authors.

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Competing interests. The authors declare that they have no conflict of interest.

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