General comments:

In this study remote satellite derived PM2.5 fields are presented. These fields have been obtained based on GOCI observations of AOD and in-situ ground-based PM2.5

- 5 measurements that in turn has been linked to each other according to relationships between these two parameters that have been established from model simulations, where emissions inventory for China and surrounding East Asia regions have been used. I think this is a very good approach in an attempt to monitor PM2.5 from space over the present very interesting regions. However, there are two important questions or major comments that are in dispute and
- 10 must be settled before this study can be accepted for publication in ACP.

RESPONSE: We are grateful to the referee for recognizing the value of this work. Thank you for the comments and suggestions below. We have taken them into full consideration when producing the revised version of manuscript. Reponses to these comments are

15 provided below.

Major comments:

1. Second paragraph of Section 3.2 and corresponding Figure 4. It is clear that a seasonal variation in ground level PM2.5 appears for the eastern China region, and then I do not

- 20 understand why annual averages of PM2.5 are presented in Figure 4. I suggest to subdivide the comparison between GOCI-derived and ground measured PM2.5 with respect to season. I suggest also to present statistic results of for example relative RMSE when comparing satellite and ground-based PM2.5.
- RESPONSE: Thanks for the suggestion. The annual averages of PM_{2.5} presented in Fig. 4 are useful for the assessment of PM_{2.5} exposure (i.e., estimation of disease burden from PM2.5 by the World Health Organization). We have added the seasonal comparison between GOCI-derived and ground measured PM_{2.5} next to the annual comparison in Fig. 4. We have also revised the related text from page 13 line 18 to page 14 line 3. Relative RMSE
 is now presented in Fig. 2, Fig. 5 and Fig. 6.
 - 1

2. The language is on the whole confusing and need to be improved, language is clearer in some few chapters and less clear in most others. Some suggestions are presented in "Specific comments" below, however, the full text needs an English proof-check.

5 RESPONSE: Thanks for the suggestion and helpful comments in "Specific comments". We have carefully revised the language and have modified the text according the "Specific comments" below.

Minor comments

1. Page 5, Lines 10-13. Clarify if you mean comparison to polar satellites? The temporal resolution is indeed higher for the present platform, but then with respect to the time period 9:00 - 16:00, which should be pointed out in the text. How is the factor 8 estimated?

RESPONSE: The factor 8 was estimated by considering the fact that traditional low-Earth polar-orbiting satellite instruments usually provide 1 retrieval/day over a location, whereas GOCI provides 8 hourly retrievals/day. On Page 4 line 9-12, we have changed to "GOCI has a high observation density of 8 retrievals/day (hourly retrievals from 09:00 to 16:00 Korean Standard Time) over a location, which exceeds the retrieval density of traditional low-Earth polar-orbiting satellite instruments".

20

2. Section 2.2. AERONET level 2 is to prefer, are these data not available for 2013?

RESPONSE: AERONET level 2 data for 2013 are not available for some stations discussed in this paper. Thus, we use level 1.5 data for consistency. We have also compared level 2 and
level 1.5 data for 2013 for stations that do have level 2 available, and found level 1.5 AOD is highly consistent with level 2 AOD with RMSE of 0.01-0.02 (relative RMSE of 2%-7%).

3. Considering the statistical equations (1 - 3) on page 10. I do not see any reason to use eq. 1 without presenting relative RMSE, and neither eq. 2 if the skewness of the deviation is not

30 discussed in the manuscript. For example instead of presenting RMSE of 0.079 (however, keep this value in the figure) at line 7 on page 11 focus on presenting the relative RMSE when you discuss the results. What is meant by a "forecast value" at line 9 on page 10? I suppose you do not deal with predictions in this work.

RESPONSE: We have added the definition of relative RMSE in Section 2.5 and have focused on relative RMSE in result discussions. We have included discussions about the skewness of the deviation when MFE or MFB is discussed. We have also changed the "forecast value" to "satellite-derived value" and substituted the "F_i" in equation to "S_i".

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4. Page 11, Lines 2 -3: Clarify if the 10.3% is obtained for this special case or for all data of 2013 investigated in the study?

RESPONSE: We have changed to "Our filters reject 10.3% of all the operational GOCI 10 AOD data investigated in this study".

5. Page 11, Lines 14-16: Then it seems that you do not have any advantages with the higher temporal resolution (minor comment 1 above) when you use the GOCI retrievals in the present investigation area, or thus the results have some other implications?

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RESPONSE: The results have other implications as suggested. The strength of higher temporal resolution lies in data density. Thus, on page 14 line 4-7, we clarified this implication by adding "we also estimated $PM_{2.5}$ from MODIS Collection 6 AOD for 2013, and found GOCI-derived $PM_{2.5}$ achieves greater consistency than MODIS-derived $PM_{2.5}$ when compared with ground based measurements (clope=1.1, $r^2=0.61$)."

20 when compared with ground-based measurements (slope=1.1, r²=0.61)."

6. Pages 12-13 and paragraph beginning with "Figure 4 compares…" Based on the results in Figure 2 left bottom I suppose several relationships between AOT and PM2.5 have been obtained with respect to seasons with GEOS-chem model? If this is not mentioned clearly in

25 the manuscript you have to do that. As it is written here and presented in the figure, I suppose the correlation coefficient of 0.81 is obtained based on the fitted linear curve with slope 1.01. You should then instead present the squared correlation coefficient (coefficient of determination), since R2 describe how well the fitted curve explain variance divided by the total variance. Similar for MODIS derived PM2.5.

30

RESPONSE: We have changed the text on page 8 line 18 to "We apply GEOS-Chem to simulate daily relationships between ground level PM_{2.5} and column AOD, specifically PM2.5/AOD" to make clear that the simulated relationship is on a daily basis. We have also

replaced the correlation coefficient with the squared correlation coefficient in Fig. 4 and related text.

7. Page 14, sentence beginning on line 16: I guess, based on the results in Figure 2 (left top
and left bottom figures), that the seasonal variation in mixing height is the main factor
explaining the variations in PM2.5 shown in Figure 6?

RESPONSE: Mixing height, emissions and nitrate formation all play a role. The topic sentence of Fig. 6 has been rephrased to emphasize that point. Fig. 2 also reflects those three processes. We also offer the mixing height change over northeastern China to more easily relate with Fig. 2.

Specific comments

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Page 2, line 6: You have to write out AERONET here, since the abstract is separated from the
main text in the manuscript. In the same way you do not need define PM2.5 and AOD once
again in Section 4, since this section is not separated from the previous sections where you
already have written out the names of these two parameters.

RESPONSE: Thanks for your advice. We have written out AERONET in the abstract and removed the definitions of PM_{2.5} and AOD in Section 4.

Page 3, line 7: Write put units after 500.

RESPONSE: We have removed the sentence containing 500 in the revised manuscript
before uploading to ACPD, because the unit is confusing and this statement is not important. The initial 500 was AQI (air quality index) of PM_{2.5} which is unitless.

Line 8: "Thus, it is....."

30 **RESPONSE:** We have corrected it in the revised manuscript before uploading to ACPD.

Line 9: Suggestion "remote sensing has a high potential to monitor PM2.5."

RESPONSE: Revised.

Line 12: Suggestion "have long been recognized to relate to ground level PM2.5...."

5 **RESPONSE:** We have changed it in the revised manuscript before uploading to ACPD.

Lines 14-15: Suggestion "....surface PM2.5 to estimate surface......"

RESPONSE: We have changed to "Many studies have developed advanced statistical relationships to estimate with high accuracy surface PM_{2.5} from satellite AOD."

Lines 17-18: The word "significant" does not suit here. "...and found relatively good correlation (R2=0.64)"

15 **RESPONSE: Revised.**

Line 22: ".....demonstrated using data from the Multiangle...."

RESPONSE: We have changed it in the revised manuscript before uploading to ACPD.

20

Page 4, lines 6-7: "...to produce a 15-year (1998-2012) global trend in ground-level PM2.5."

RESPONSE: We have changed it in the revised manuscript before uploading to ACPD.

Line 18: "speciation" ?

RESPONSE: We have replaced "speciation" with "composition" for the entire manuscript.

Line 19: "of the current"

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RESPONSE: Revised.

Page 5, line 8: Change to "6 km2"

RESPONSE: We have changed to "6km by 6km" for the entire manuscript.

Lines 14-18: Suggestion "A challenge using GOCI to detect aerosols in the atmosphere is the absence of mid-infrared.....to detect clouds, which means that significant errors are probably induced in the estimates of AOD. The operational product screen clouds based on spatial......at each pixel in combination with a......at 4km2 resolution aboard......."

RESPONSE: We have changed it in the revised manuscript before uploading to ACPD.

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Line 19: "However, as will be shown here cloud......"

RESPONSE: We have changed it in the revised manuscript before uploading to ACPD.

15 Lines 21-22: The filter methods include 1) setting a minimum number of 15 valid AOT values per grid cell of 30 km2, 2) using a local variance check to eliminate grid cells where the coefficient...... of AOD is larger than 0.5 within a grid cell, and 3)....."

RESPONSE: We have changed it in the revised manuscript before uploading to ACPD.

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Page 6, Line 5: "ground-based"

RESPONSE: We have replaced all the "ground measurements" to "ground-based measurements" in the revised manuscript before uploading to ACPD.

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Lines 8-9, "Here we use AERONET level 1.5 cloud screened data (Smirnov et al., 2000) from 4 stations within the GOCI domain: Beijing.....and EPA-NCU."

RESPONSE: Revised.

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Lines 11-12: "Criteria for selecting an AERONET station is 1) a PM2.5 ground-based station has to be located within 10 km and 2) a complete time series of PM2.5 data for the period of study has to be available.

RESPONSE: We have changed it in the revised manuscript before uploading to ACPD.

Line 14: "However, due to interrupted time series of PM2.5 at both these stations we

5 combine the AERONET AOD from the Beijing and Beijing-CAMS stations and PM_{2.5} from the corresponding two *in-situ* ground-based sites."

RESPONSE: Revised.

Authors' Response to Comments from Referee #3

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We would like to thank Referee 3 for his/her useful comments and suggestions that helped to improve the quality of this manuscript. Reponses to these comments are provided below.

Specific Comments:

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1) Page 17254, Introduction:

Suggest have one paragraph describing why using the Korean Geostationary Ocean Color Imager (GOCI) to quantity surface PM2.5 concentrations in the eastern China. As the authors mentioned, this approach has been applied to a number of satellite

- 20 instruments: MODIS, MISR, and SeaWiFS. What are the advantages to use the GOCI instrument?
 - RESPONSE: Thanks for the suggestion. The advantage of GOCI instrument lies in high data density due to geostationary nature, which enables more detailed investigation on aerosol properties at regional scale. We have added the following
 - paragraph on page 4 line 8-14 to highlight this advantage.

"The Geostationary Ocean Color Imager (GOCI) is the first geostationary satellite instrument that offers multi-spectral aerosol optical properties in Northeast Asia

30 (Park et al., 2014). GOCI has a high observation density of 8 retrievals/day (hourly

retrievals from 09:00 to 16:00 Korean Standard Time) over a location, which exceeds the retrieval density of traditional low-Earth polar-orbiting satellite instruments. Thus, GOCI is promising for more detailed investigations on aerosol properties in highly polluted and populated regions including eastern China."

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2) Page 17256, Section 2.1:

In the manuscript, the authors emphasized the importance of cloud filters at a number of places. How sensitive are the conclusions to the three cloud filters? For example, the first filter set a minimum number of 15 retrievals per 30km x 30km grid, how would the

10 results change if using the number of 10 retrievals?

RESPONSE: Thanks for the suggestion. The thresholds of filters described in this manuscript actually present the best consistency between satellite data and ground-based observations. For example, changing the threshold of buddy check in our

- 15 cloud filters from 15 to 10 would significantly underestimate AOD especially in northern Taiwan where the MFB would increase from -1.2% to -15.0%. Limiting the diurnal variation of GOCI AOD to the 80th percentile of diurnal variations in observations would introduce bias (rRMSE would increase by 4% in Beijing) in GOCI AOD. Decreasing the threshold of local variance check to 0.4 has little
- 20 influence (< 0.1% for all statistics) on AOD, but would introduce bias to GOCI-derived PM_{2.5} by depressing the variation of PM_{2.5} concentrations. This is reflected by the underestimated GOCI-derived PM_{2.5} in Beijing areas (rRMSE=16.6% and MFB=-6.8%) and Shandong areas (rRMSE=16.6% and MFB=-3.42%) where PM_{2.5} concentrations are higher than average (Table 1). We have added this description to page 12 line 5-12 and page 14 line 18-23 to clarify this point.

4) Page 17258, Line 5:

It appears that Heald et al. (2012) has tested a few ways to correct the HNO3 overestimates over the United States. Can you describe which ones you have

30 implemented in your study? And does the HNO3 overestimation also apply over the eastern China?

RESPONSE: In model, we reduced HNO₃ concentrations at each timestep to 75% of their original values to correct the overestimation. The HNO₃ overestimation is also found over eastern China by Wang et al. (2013). We have revised page 8 line 14-15

5 to describe the implementation of this HNO₃ correction.

References:

Wang, Y., Zhang, Q. Q., He, K., Zhang, Q. and Chai, L.: Sulfate-nitrate-ammonium aerosols over China: response to 2000–2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia, Atmos. Chem. Phys., 13(5), 2635–2652, doi:10.5194/acp-13-2635-2013, 2013.

5) Page 17262, Line 1-4:

It would be helpful to explain the comparisons with GEOS-Chem and MODIS-derived PM2.5 concentrations. Are these your results or from previous studies?

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RESPONSE: These are our results. We have revised the text to "Using the same technique, we also estimated $PM_{2.5}$ from MODIS Collection 6 AOD for 2013, and found GOCI-derived $PM_{2.5}$ achieves greater consistency than MODIS-derived $PM_{2.5}$ when compared with ground-based measurements (slope=1.1, r²=0.61)" on page 14 line 4-7.

6) Page 17262, Section 3.3:

This section discussed the chemical speciation of satellite-derived PM2.5. I suggest add a few more sentences describing how you derived the chemical composition of satellite-

25 derived PM2.5 and a new figure showing their spatial distribution (like the panel of Figure 3). These would help to support the discussion here.

RESPONSE: Thanks for the suggestion. We firstly used GEOS-Chem to simulate an annual mass fraction of chemical components in PM_{2.5}, and then applied the

30 simulated mass fraction to GOCI-derived PM_{2.5} concentration to estimate the mass



concentrations of components in GOCI-derived PM_{2.5}. We have modified the text to "Fig. 6 also shows the chemical composition of GOCI-derived PM_{2.5}, as calculated by applying the GEOS-Chem simulated mass fraction of PM_{2.5} chemical components to GOCI-derived PM_{2.5} mass concentration" on page 15 line 6-8.

5 We have also provided the spatial distribution of GOCI-derived PM_{2.5} chemical composition and related text in supplementary materials.

7) Page 17263, Section 3.3:

Is there any difference between Organic Matter (OC) and Organic Carbon (OC)?

10 Please clarify.

15

RESPONSE: Yes, organic matter (OM) is constituted of organic carbon (OC) as its major component and other element such as hydrogen, oxygen and nitrogen. We have clarified the difference by adding "OM, which includes elements such as hydrogen, oxygen and nitrogen" to page 7 line 19-20.

8) Page 17265, Line 9-14:

Please tell us how you estimated the health impact. By summing up the population over areas with PM2.5 concentration above 35 ug m-3? From Figure 3, it did not seem to me

20 that all regions of eastern China exceed 35 ug m-3 (with the color scale goes to zero).

RESPONSE: The population of eastern China in Table 1 was a simple sum of population geographically located in eastern China. As the referee implied, a more accurate way to estimate the health impact is to sum up the population over areas

25 where PM_{2.5} concentration exceeds 35 ug m⁻³. Therefore, we have changed the "Population (million people)" row of Table 1 to "Population (million people) exposed to PM_{2.5} exceeding IT-1 level", and re-calculated the population. We have also changed the text on page 18 line 11-15 accordingly. Besides, we modified the colormap of Fig. 3 to provide clearer distribution when PM_{2.5} concentrations are
30 below 50 ug m⁻³.

9) Page 17275, Table 1:

Please clarify whether the concentrations of different chemical speciation are populationweighted or area-weighted.

5 **RESPONSE:** The concentrations of chemical composition are area-weighted. We modified the caption of Table 1 to specify this point.

Technical Comments:

10 1) Page 17253, Line 2:

East China or the eastern China? Please be consistent. The term East China represents a specific geographic domain.

RESPONSE: Thanks. We have substituted all East China to eastern China in text.

15

2) Page 17254, Line 27:

Please define the domain of the eastern China in the study, the domain of Figure 3 or by longitude and latitude?

20 RESPONSE: We added country borders to Fig. 3 to identify eastern China.

3) Page 17256, Line 7:

Please define mathematically the coefficient of variation, probably in Section 2.5.

25 **RESPONSE:** We added the mathematical equation of coefficient of variation in Section 2.5.

4) Page 17260, Line 19:

Suggest change the forecast value to the model simulated value

30

RESPONSE: Thanks. We have revised the "forecast value" to "satellite-derived value" and have substituted the " F_i " in equation to " S_i "

Estimating Ground-level PM_{2.5} in Eastern China Using Aerosol

Optical Depth Determined from the GOCI Satellite Instrument

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Abstract

We determine and interpret fine particulate matter (PM_{2.5}) concentrations in eastern China for January to December 2013 at a horizontal resolution of 6 km from aerosol optical depth (AOD) retrieved from the Korean Geostationary Ocean Color Imager
(GOCI) satellite instrument. We implement a set of filters to minimize cloud contamination in GOCI AOD. Evaluation of filtered GOCI AOD with AOD from the Aerosol Robotic Network (AERONET) indicates significant agreement with mean fractional bias (MFB) in Beijing of 6.7% and northern Taiwan of -1.2%. We use a global chemical transport model (GEOS-Chem) to relate the total column AOD to the near-

- 10 surface PM_{2.5}. The simulated PM_{2.5}/AOD ratio exhibits high consistency with ground-based measurements in Taiwan (MFB= -0.52%) and Beijing (MFB= -8.0%). We evaluate the satellite-derived PM_{2.5} versus the ground-level PM_{2.5} in 2013 measured by the China Environmental Monitoring Center. Significant agreement is found between GOCI-derived PM_{2.5} and *in-situ* observations in both annual averages (r²=0.66, N=494) and monthly averages (relative RMSE=18.3%), indicating GOCI provides valuable data for air quality studies in Northeast Asia. The GEOS-Chem simulated chemical composition of GOCI-derived PM_{2.5} reveals that secondary inorganics (SO₄²⁻, NO₃⁻, NH₄⁺) and organic matter are the most significant components. Biofuel emissions in northern China
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for heating increase the concentration of organic matter in winter. The population-

weighted GOCI-derived PM_{2.5} over eastern China for 2013 is 53.8 µg m⁻³, with 400 million residents in regions that exceed the Interim Target-1 of the World Health Organization.

1. Introduction

- 5 Fine particulate matter with aerodynamic diameter less than 2.5 µm (PM_{2.5}) is a robust indicator of mortality and other negative health effects associated with ambient air pollution (Goldberg et al., 2008; Laden et al., 2006). It is estimated that more than three million people lost their lives prematurely due to PM2.5 in 2010 (Lim et al., 2012), of which one million occurred in East Asia (Silva et al., 2013). In China, there have already
- 10 been several episodes with PM2.5 described as "beyond index" levels. Thus, it is of paramount importance to monitor PM2.5 concentration across China. Satellite remote sensing has a high potential to monitor PM_{2.5},

Satellite retrievals of aerosol optical depth (AOD), which provide a measure of the amount of light extinction through the atmospheric column due to the presence of 15 aerosols, have long been recognized to relate to ground level PM2.5 (Wang and Christopher, 2003). Many studies have developed advanced statistical relationships to estimate with high accuracy surface PM_{2.5} from satellite AOD (Liu et al., 2009; Kloog et al., 2012; Hu et al., 2013). For example, Ma et al. (2014) estimated PM₂₅ concentrations in China from satellite AOD by developing a national-scale geographically weighted regression model, and found strong agreement ($r^2=0.64$) with ground measurements.

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In addition to empirical statistical methods, satellite AOD can also be geophysically related to surface PM2.5 by the use of a chemical transport model to simulate the PM2.5 to

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AOD relationship (Liu et al., 2004; van Donkelaar et al., 2010). This approach was first demonstrated using data from the Multiangle Imaging Spectroradiometer (MISR) aboard NASA's Terra satellite over the United States for 2001 (Liu et al., 2004). Van Donkelaar et al. (2006, 2010) extended this approach to estimate PM2.5 from AOD retrieved from

- 5 both the MODIS (Moderate Resolution Imaging Spectroradiometer) and the MISR satellite instruments, and developed a long-term global estimate of PM2.5 at a spatial resolution of approximately 10 km x 10 km. Boys et al. (2014) used AOD retrieved from MISR and the SeaWiFS (Sea-viewing Wide Field-of-view Sensor) to produce a 15-year (1998-2012) global trend of ground-level PM2.5. These previous studies have proven to
- 10

polluted and populated regions like China.

The Geostationary Ocean Color Imager (GOCI) is the first geostationary satellite instrument that offers multi-spectral aerosol optical properties in Northeast Asia (Park et al., 2014). GOCI has a high observation density of 8 retrievals/day (hourly retrievals from

be globally effective, but more detailed regional investigation is needed in densely

15 09:00 to 16:00 Korean Standard Time) over a location, which exceeds the retrieval density of traditional low-Earth polar-orbiting satellite instruments. Thus, GOCI is promising for more detailed investigations on aerosol properties in highly polluted and populated regions including eastern China.

In this study, we estimate ground-level PM_{2.5} in eastern China for 2013 at a horizontal 20 resolution of 6 km by 6 km, by using AOD retrieved from GOCL coupled with the relationship of PM_{2.5} to AOD simulated by a chemical transport model (GEOS-Chem). Section 2 describes the approach and data, Section 3 evaluates the GOCI AOD, the

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simulated $PM_{2.5}$ to AOD relationship, and the GOCI-derived $PM_{2.5}$ using recently available ground-level measurements from the China Environmental Monitoring Center (http://113.108.142:20035/emcpublish/). We also interpret the GOCI-derived $PM_{2.5}$ by using the GEOS-Chem model to estimate its chemical <u>composition</u>, Section 4 summarizes the major findings and potential future improvements <u>of</u> the current analysis.

2. Methods

2.1 Aerosol optical depth from the GOCI satellite instrument

GOCI operates onboard the Communication, Ocean, and Meteorology Satellite (COMS) that was launched in 2010 in Korea (Lee et al., 2010). The spatial coverage of GOCI is

2500 km x 2500 km in Northeast Asia, including eastern China, the Korean peninsula and Japan (Kang et al., 2006). GOCI has eight spectral channels for aerosol retrievals, including six visible bands at 412, 443, 490, 555, 660, 680 nm and two near infrared bands at 745 and 865 nm (Park et al., 2014). The Level 2 AOD products are retrieved at a spatial resolution of 6_km_by_6 km, using a clear-sky composite method for surface reflectance and a lookup table approach based on AERONET observations (Lee et al.,

2010; Lee et al., 2012).

A challenge using GOCI to detect aerosols in the atmosphere is the absence of midinfrared (IR) channels to detect clouds, which means that significant errors could be induced in the estimates of AOD. The operational GOCI products screen clouds based on

spatial variability and threshold tests at each 6 km x 6 km pixel in combination with a meteorological imager that has 4 IR channels (at 3.7 μm, 6.7 μm, 10.8 μm, 12 μm wavelengths) at 4 km by 4 km resolution onboard the same satellite (Cho et al., 2006).

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optical properties in Northeast Asia.

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Deleted: From its geostationary platform, GOCI has the capacity to provide hourly aerosol optical properties from09:00 to 16:00 Korean Standard Time (Park et al., 2014). However, as will be shown here cloud contamination still occurs. Therefore, we apply a set of spatial filters following Hyer et al. (2011) and temporal filters to further eliminate cloud contamination in GOCI AOD. The filters include 1) a buddy check that sets a minimum number of 15 retrievals per 30 km x 30 km grid cell, 2), a local variance check

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to eliminate grid cells where the coefficient of variation of AOD is larger than 0.5 within the <u>surrounding 5 x 5 grid cells</u> and 3) a diurnal variation check that excludes grid cells with diurnal variation (maximum – minimum) of AOD larger than 0.74 which is the 90th percentile of diurnal variation of AERONET AOD in Beijing and northern Taiwan for 2013. In this study, we use GOCI AOD for Jan-Dec 2013 to derive ground-level PM2.5 in

10 eastern China.

2.2 Aerosol optical depth from AERONET ground-based measurements

The Aerosol Robotic Network (AERONET) is a globally distributed network of CIMEL Sun photometers (Holben et al., 1998) that provide multi-wavelength AOD measurements with a low uncertainty of < 0.02 (Holben et al., 2001). Here we use AERONET Level 1.5 cloud screened data (Smirnov et al., 2000) for Jan-Dec 2013 from 4 stations within the GOCI domain: Beijing, Beijing-CAMS, Taipei CWB and EPA-NCU. Criteria for selecting an AERONET station are 1) a PM_{2.5} ground monitor has to be located within 10 km and 2) a complete time series of AOD data records for the period of study has to be available. Beijing and Beijing-CAMS stations are located in downtown 20 Beijing, with the closest available PM_{2.5} monitors 9.5 km and 7.5 km away, respectively. However, due to interrupted time series of PM_{2.5} records at both these stations, we combine the AERONET AOD from the Beijing and Beijing-CAMS stations and PM_{2.5} from the corresponding two *in-situ* ground-based sites as a "combined Beijing" site.

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Taipei_CWB and EPA-NCU <u>stations</u> are located in populated northern Taiwan, with nearly collocated $PM_{2.5}$ monitors (< 3 km). We similarly combine the Taipei_CWB and EPA-NCU as "northern Taiwan" site. We use these sites to evaluate GOCI AOD and the relationship between AOD and $PM_{2.5}$ simulated by a global chemical transport model.

5 2.3 Simulation of the relationship between AOD and PM_{2.5} by GEOS-Chem

We use the GEOS-Chem chemical transport model (version 9-01-03; http://geoschem.org) to calculate the spatiotemporally resolved relationship between ground-level PM_{2.5} and satellite-retrieved column AOD.

Our nested GEOS-Chem simulation at 1/2° x 2/3° spatial resolution with 47 vertical 10 levels (14 levels in the lowest 2 km) is driven by assimilated meteorology from the Goddard Earth Observing System (GEOS-5). A global simulation at 2° x 2.5° spatial resolution is used to provide boundary conditions for the nested domain (Wang et al., 2004). We spin up the model for one month before each simulation to remove the effects of initial conditions on the aerosol simulation.

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GEOS-Chem includes a fully coupled treatment of tropospheric oxidant-aerosol chemistry (Bey et al., 2001; Park et al., 2004). The GEOS-Chem aerosol simulation includes the sulfate-nitrate-ammonium system (Park et al., 2004; Pye et al., 2009), primary (Park et al., 2003) and secondary (Henze et al., 2006; Henze et al., 2008; Liao et al., 2007; Fu et al., 2008) organics, mineral dust (Fairlie et al., 2007), and sea salt (Jaegle et al., 2011). We estimate the concentration of organic matter (OM, which includes elements such as hydrogen, oxygen and nitrogen) from the simulated primary organic

carbon (OC) using spatially and seasonally <u>resolved</u> values from OMI (Ozone Monitoring Instrument) NO₂ and AMS (Aerosol Mass Spectrometer) measurements following Philip et al. (2014). Gas-aerosol phase partitioning is simulated using <u>the</u> ISORROPIA II thermodynamic scheme (Fountoukis and Nenes, 2007). GEOS-Chem calculates AOD using relative humidity dependent aerosol optical properties following Martin et al.

(2003). Dust optics are from Ridley et al. (2012).

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Anthropogenic emissions are based on the Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org) for 2010, and the Zhang et al. (2009) inventory for

- 10 surrounding East Asia regions for 2006. Both inventories are scaled to the simulation year (2012-2013), following Ohara et al. (2007). Non-anthropogenic emissions include biomass burning emissions (GFED-3) (Mu et al., 2011), biogenic emissions (MEGAN) (Guenther et al., 2006), soil NO_x (Yienger and Levy, 1995; Wang et al., 1998), lightning NO_x (Murray et al., 2012), aircraft NO_x (Wang et al., 1998; Stettler et al., 2011), ship SO₂
- from EDGAR (Olivier et al., 2001) and volcanic SO₂ emissions (Fischer et al., 2011).
 <u>HNO₃ concentrations are artificially decreased to 75% of their values at each timestep</u>
 <u>following Heald et al. (2012) to account for regional bias (Wang et al., 2013).</u> Emissions are distributed into the lower mixed layer, with a correction to the GEOS-5 predicted nighttime mixing depths following Heald et al. (2012) and Walker et al. (2012).
- We apply GEOS-Chem to simulate <u>daily</u> relationships between ground level $PM_{2.5}$ and column AOD, specifically $PM_{2.5}$ /AOD. $PM_{2.5}$ concentrations are calculated at 35% relative humidity for consistency with <u>in-situ</u> measurements. For consistency with GOCI AOD and $PM_{2.5}$ ground-based measurements, we sample the simulated AOD only from
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hours that GOCI has retrievals (00:00 - 07:00 UTC), and calculate the simulated daily $PM_{2.5}$ from 24-hour averages as reported for the ground-based $PM_{2.5}$ measurements. The simulation period is May 2012 - April 2013 as the GEOS-5 meteorological fields are not available afterward. The mismatch with observations for May-Dec 2013 has the potential to degrade performance, but as will be shown here no clear loss of quality is apparent.

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2.4 <u>In-situ</u> PM_{2.5} measurements

We collect $PM_{2.5}$ measurements from 494 monitors to evaluate the GOCI-derived values. <u>*In-situ*</u> $PM_{2.5}$ daily measurements in Mainland China for 2013 are primarily from the official website of the China Environmental Monitoring Center (CEMC; <u>http://113.108.142:20035/emcpublish/</u>). Data are also collected from some provinces (e.g.

- Shandong, Zhejiang) and municipalities (e.g. Beijing and Tianjin) with additional sites that are not included in the CEMC website. Daily *in-situ* PM_{2.5} data in northern Taiwan for 2013 are from the Taiwan Environmental Protection Administration (TEPA; http://taqm.epa.gov.tw). The *in-situ* PM_{2.5} data in both Mainland China and northern
- Taiwan are measured by a collection of the Tapered Element Oscillating Microbalance Methods (TEOMs) and beta-attenuation methods (BAMs) with some TEOMs being heated to 30 °C and others to 50 °C (CNAAQS GB3095-2012, 2012; http://taqm.epa.gov.tw). The specific instrument (BAMs or TEOMs) used by each monitoring site is unknown. The effective relative humidity of the resultant PM_{2.5}
 measurement likely varies diurnally and seasonally as a function of the ambient temperature. Semivolatile losses are expected from the TEOMs. The network design appears to include compliance objectives that may affect monitor placement. Despite

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these issues, we use the monitoring data to evaluate our satellite-derived PM_{2.5} since the

monitoring data offer valuable information about ground-level $PM_{2.5}$ concentrations. We also collect $PM_{2.5}$ measurements from a monitor in Beijing <u>as part of the Surface</u> PARTiculate mAtter Network (SPARTAN; <u>www.spartan-network.org</u>) using a three-wavelength nephelometer and an impaction filter sampler (Snider et al., 2015). The

5 SPARTAN, CEMC and TEPA PM_{2.5} monitoring data combined with AERONET AOD are used to estimate the empirical relationship between PM_{2.5} and AOD, and to further evaluate the relationship simulated by the model.

2.5 Statistical Terms

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Root mean square error (RMSE), <u>relative root mean square error (rRMSE)</u>, mean fractional bias (MFB) and mean fractional error (MFE) are defined as

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (S_i - O_i)^2}$$
(1)

$$\underline{\mathrm{rRMSE}} = \frac{\mathrm{RMSE}}{\frac{1}{N} \sum_{i=1}^{N} 0_i}$$
(2)

$$MFB = \frac{1}{N} \sum_{i=1}^{N} \frac{(S_i - O_i)}{(\frac{S_i + O_i}{2})} \times 100\%$$
(3)

MFE =
$$\frac{1}{N} \sum_{i=1}^{N} \frac{|s_i - 0_i|}{(\frac{S_i + 0_i}{2})} \times 100\%$$
 (4)

Coefficient of variation (CV) is defined as

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3. Results and Discussion

3.1 Evaluation of satellite AOD and the simulated relationship between PM2.5 and AOD

Figure 1 shows the effects of our cloud-screening filters on GOCI AOD. The left panel shows GOCI true color images from 5 July 2013 at 10:30 (top) and 11:30 (bottom)

- 5 Korean Standard Time. The boxes identify challenging regions with thick white cloud, dark cloud-free oceans and grey shading that appears to be thin cloud. The operational GOCI AOD retrievals, shown in the middle panel, correctly exclude thick clouds, but report high AOD for the potentially thin clouds. Although these grey regions could contain aerosol, we err on the side of caution. Application of our additional temporal and
- 10 spatial cloud filters removes the suspicious pixels from the original GOCI data, as shown in the right panel. Our filters reject 10.3% of <u>all</u> the operational GOCI AOD data investigated in this study. We evaluate the cloud filters further below.

Figure 2 (top) shows monthly averages of coincident filtered hourly GOCI and AERONET AOD for Jan-Dec 2013 at combined Beijing and northern Taiwan stations.

 GOCI AOD is highly consistent with AERONET observations with MFB of 6.7% in Beijing and -1.2% in Taiwan. GOCI AOD and AERONET AOD are positively skewed at both stations, and the skewness is reduced in GOCI AOD at both stations due to more records for extremely small AOD (< 0.04) in GOCI products. The relatively larger rRMSE between GOCI and AERONET AOD in northern Taiwan may reflect the fewer
 observations there.

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We investigate the filtered diurnal variation of GOCI AOD at the above AERONET stations and find the level of AOD is uniform within a day (e.g. the coefficient of variation in Beijing is 0.1), similar to AERONET observations.

The effect of excluding our cloud-screening filters is negligible for coincident

- 5 comparisons with AERONET since AERONET is already cloud-screened. The exclusion of our cloud filters for a non-coincident comparison that includes all GOCI data would introduce significant error versus AERONET observations, increasing <u>rRMSE</u> by a factor of 1.7, 3, 3 in Beijing and northern Taiwan. Changing the buddy check threshold in our cloud filters from 15 to 10 would significantly underestimate AOD especially in northern
- 10 Taiwan where the MFB would increase from -1.2% to -15.0%. Decreasing the threshold of local variance check to 0.4 has little influence (< 0.1%), for rRMSE, MFB and MFE, but would have larger influence on GOCI-derived $PM_{2.5}$ as will be shown later. Limiting the diurnal variation of GOCI AOD to the 80th percentile of diurnal variations in observations would introduce bias (rRMSE would increase by 4% in Beijing) to GOCI

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15 <u>AOD.</u> As will be shown here, GOCI-derived PM_{2.5} offers an additional test of cloud screening filters.

Figure 2 (bottom) shows the relationship between the ground level PM_{2.5} and the columnar AOD as simulated by GEOS-Chem and from ground-based measurements. The measured ratio in Beijing has pronounced seasonal variation with values high in winter and low in spring. The measured ratio in northern Taiwan exhibits little seasonal variation. The annual mean GEOS-Chem PM_{2.5}/AOD ratio well reproduces the ground-

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based measurements despite the temporal inconsistency of the two metrics for May - Dec. The simulation captures the pronounced seasonal variation in Beijing and the comparably

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aseasonal behavior in northern Taiwan. The simulated seasonal variation of PM2.5/AOD in Beijing arises from the seasonal variation of mixed layer depth (factor of 2 higher in summer than winter) combined with the near-constant columnar AOD throughout the year as shown in Fig. 1 (top).

5 Snider et al. (2015) interpreted coincident measurements of AOD, PM2.5, and nephelometer measurements of aerosol scattering and found that the temporal variation of the PM_{2.5}/AOD ratio in Beijing was primarily driven by the vertical profile in aerosol scattering. We examine the seasonal variation in the simulated $PM_{2.5}/AOD$ and similarly find that the ratio of ground-level aerosol scatter, to columnar AOD contributes most (89%) of the monthly variability in the PM_{2.5}/AOD ratio in Beijing.

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3.2 Evaluation of ground-level PM2.5 derived from GOCI AOD

Figure 3 shows the seasonal and annual distribution of PM2.5 over East Asia at a spatial resolution of 6 km by 6 km for 2013. In both GOCI-derived and measured PM2.5, winter concentrations in eastern China exceed 100 μ g m⁻³ over vast regions, with lower values in summer. Both GOCI-derived and *in-situ* measurements reveal that PM2.5 in northern China is higher than in southern China, especially for the Beijing, Hebei and Shandong provinces where the annual PM2.5 is almost 100 µg m⁻³ or more. Prior work has attributed this regional enhancement to high emission rates (Zhao et al., 2013; Zhang et al., 2013), that in part arises from exports (Jiang et al., 2015).

20 Figure 4 compares annual and seasonal averages of daily ground-measured PM2.5 from 494 sites with coincident daily GOCI-derived PM2.5 from pixels that contain the groundbased sites. A significant correlation ($r^2=0.66$, N=494) with a slope near unity (1.01) is

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found in the annual scatter plot. The slope remains near unity (0.95-1.01) in seasonal scatter plots. The weaker correlation for all four seasons implies random representativeness differences between point *in situ* measurements and area-averaged satellite values when data density diminishes. Semivolatile losses from some *in situ* instruments (TEOMs) might contribute to scatter in winter when nitrate constitutes a larger fraction of $PM_{2.5}$. We focus on more meaningful aggregated measurements. Using the same technique, we also estimated $PM_{2.5}$ from MODIS Collection 6 AOD for 2013, and found GOCI-derived $PM_{2.5}$ achieves greater consistency than MODIS-derived $PM_{2.5}$ when compared with ground-based measurements (slope=1.1, r²=0.61). GOCI-derived

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10 $\underline{PM_{2.5}}$ also corrects the significant underestimation of $\underline{PM_{2.5}}$ from GEOS-Chem (slope=0.68, r²=0.85) when compared with ground measurements.

Figure 5 shows monthly averages of GOCI-derived $PM_{2.5}$ and <u>in-situ</u> measurements at four regions outlined in Fig. 3. Regions are selected based on the level of $PM_{2.5}$ concentration and the population of residents. A high degree of consistency is found in all

- 15 regions. Both datasets show more seasonal variation in northern regions like Beijing and Shandong than southern regions like Shanghai and northern Taiwan. Both indicate that PM_{2.5} concentrations in northern regions are generally higher than in southern regions. The exclusion of our cloud screening filters from the GOCI AOD would introduce significant bias in GOCI-derived PM_{2.5} versus ground-based measurements especially in
- 20 summer, increasing rRMSE by a factor of 1.7 5.3 in all four regions. Changing the threshold of local variance check in our cloud filters to 0.4 would introduce bias by restricting the variation of PM_{2.5} concentrations. For example, GOCI-derived PM_{2.5}

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would be generally underestimated in Beijing areas (rRMSE=16.6% and MFB=-6.8%) and Shandong areas (rRMSE=16.6% and MFB=-3.42%).

3.3 Seasonal variation of PM_{2.5}

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Figure 6 shows the monthly averages of coincident daily GOCI-derived and <u>in-situ PM_{2.5}</u> concentrations for the domain of eastern China. <u>Both the GOCI-derived PM_{2.5} and</u> ground-based observations <u>exhibit</u> similar seasonal variation with values high in winter and low in summer. Exclusion of our temporal and spatial cloud-screening filters from GOCI-derived PM_{2.5} would increase <u>rRMSE</u> by a factor of 3.4. Fig. 6 also shows the chemical <u>composition</u> of GOCI-derived PM_{2.5}, as calculated by applying the GEOS-

- 10 Chem simulated mass fraction of $PM_{2.5}$ chemical components to GOCI-derived $PM_{2.5}$ mass concentration. Aerosol water is attached to each component according to its hygroscopicity. Secondary inorganic aerosols (SIA; $SO_4^{2^2}$, NO_3^- , NH_4^+) are the most abundant components throughout the year, accounting for 65% of $PM_{2.5}$ concentrations, followed by OM (18%). The NO_3^- and OM concentrations increase by a factor of 2 in
- winter, together comprising most of PM_{2.5} (31% for NO₃⁻ and 26% for OM). Summer is predominately controlled by SIA (74%). Dust plays an important role in spring (15%) and fall (15%). Our seasonal variation of chemical composition is generally consistent with ground-based measurements in previous works across eastern China. A number of studies in Beijing, the Yangtze River delta and Pearl River delta regions all reported that
 OM and SIA are the most important components of PM_{2.5} through the year (He et al., 2001; Ye et al., 2003; Tao et al., 2012; Zhang et al., 2013). Zhang et al. (2008) showed consistent seasonal patterns in OM at 18 stations in China, with a winter maximum, and a

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summer minimum, similar to the seasonality of OM in this work. Zhang et al. (2013)

studied the chemical <u>composition</u> of $PM_{2.5}$ in Beijing and <u>found</u> the percentage of SIA in $PM_{2.5}$ is largest in summer, consistent with our result.

The seasonal variation of $PM_{2.5}$ in Fig. 6 is driven by a combination of meteorological conditions, emissions, and nitrate formation. All three processes have greater seasonal

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variation in the north than south. The mixing height over northeastern China has strong seasonal variation with summer having an average mixing height from GEOS-5 that is 1.9 times higher than in winter. The GEOS-Chem simulation reveals that the increase of OM in winter is primarily driven by biofuel emissions from burning wood, animal waste and agricultural waste (Bond et al., 2004) for heating in eastern China. The spatial distribution of biofuel emission is primarily north of the Yangze River, especially from the North China Plain. The significant contribution from biofuel emissions to the OM concentration in our work is consistent with Bond et al. (2004) who found residential biofuel emissions were responsible for ~70% of OC emissions in China. The increase of NO₃⁻ in winter in Fig. 6 is consistent with prior attribution of the increase of NO₃⁻ in

winter to the favorable formation of NH₄NO₃ at low temperatures (Wang et al., 2013).
 <u>Supplemental Figure S1 shows the spatial distribution of PM_{2.5} chemical components.</u>

Table 1 shows the annual chemical <u>composition</u> of GOCI-derived $PM_{2.5}$ in regions outlined in Fig. 3 and in overall eastern China. SIA and OM are the most abundant species. Among the SIA components, $SO_4^{2^-}$ and NO_3^- concentrations are similar in the Beijing, Shandong and Shanghai regions, whereas in eastern China and northern Taiwan $SO_4^{2^-}$ is the dominant component. OM concentrations in the Beijing and Shandong regions are considerably higher than in the other regions, similar to or even exceeding the JUNWEI XU 2015-8-16 4:37 PM Deleted: speciation JUNWEI XU 2015-10-27 2:49 PM Deleted: indicated JUNWEI XU 2015-10-27 2:49 PM Deleted: the

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of OM concentrations.

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concentrations of SO_4^{2-} and NO_3^{-} . Our estimation of $PM_{2.5}$ composition is generally consistent with *in-situ* measurements in prior studies. In Beijing, the concentrations of SIA in this work are similar to Zhang et al. (2013) who measured concentrations for 2009-2010 of $13.6 \pm 12.4 \ \mu g \ m^{-3}$ for SO_4^{2-} , $11.3 \pm 10.8 \ \mu g \ m^{-3}$ for NO_3^{-} and $6.9 \pm 7.1 \ \mu g$

- 5 m⁻³ for NH₄⁺. Our SIA concentrations in Beijing are also comparable with Yang et al. (2011) who measured concentrations for 2005-2006 of $15.8 \pm 10.3 \ \mu g \ m^{-3}$ for SO₄²⁻, 10.1 $\pm 6.09 \ \mu g \ m^{-3}$ for NO₃⁻ and 7.3 $\pm 4.2 \ \mu g \ m^{-3}$ for NH₄⁺. The OC concentration in Beijing in this work is smaller than Zhang et al. (2013) of $16.9 \pm 10.0 \ \mu g \ m^{-3}$ and Yang et al. (2011) of $24.5 \pm 12.0 \ \mu g \ m^{-3}$. In Shandong and surrounding regions, our concentrations are
- smaller than in Cheng et al. (2011) by a factor of about 2, perhaps related to unresolved sources. Our results in Shanghai cluster are comparable with Yang et al. (2011) for 1999-2000, except the OC concentration in this work is considerably lower than Yang et al. (2011) of 16.8 μg m⁻³. In northern Taiwan, our NO₃⁻ is similar to Fang et al. (2002) for 2001-2003, yet our estimations of SO₄²⁻ and NH₄⁺ are higher than Fang et al. (2002) by a
- 15 factor of two, which could be driven by changes in emissions over the last decade. In summary, the chemical <u>composition</u> broadly represents <u>in-situ</u> measurements with some location-dependent discrepancies.

3.4 Population exposure to ambient PM_{2.5} in eastern China

We estimate the population exposure to ambient PM_{2.5} in eastern China for 2013 at a 20 spatial resolution of 6 km by 6 km using our GOCI-derived PM_{2.5} and the Gridded Population of the World (GPW; Tobler et al., 1997) data for 2010 from the Socioeconomic Data and Applications Center (GPW version 3; JUNWEI XU 2015-8-16 5:04 PM Deleted: in situ

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http://sedac.ciesin.columbia.edu/). Table 1 also provides the population-weighted GOCIderived $PM_{2.5}$ for regions outlined in Fig. 3 and for overall eastern China. The population-weighted $PM_{2.5}$ exceeds the area-weighted for all regions except northern Taiwan and Shandong and surrounding regions. The overall population-weighted $PM_{2.5}$

- 5 concentration for eastern China for 2013 is 53.8 μ g m⁻³. The level of PM_{2.5} for Beijing and Shandong regions in this study is similar to Ma et al. (2014) who suggested that the PM_{2.5} concentration over the North China Plain for 2013 is 85 - 95 μ g m⁻³. The PM_{2.5} concentration in eastern China in this study is also comparable with previous works. Van Donkelaar et al. (2015) estimated the PM_{2.5} concentration over eastern Asia for 2001-
- 10 2010 is $50.3 \pm 24.3 \ \mu g \ m^{-3}$. Geng et al. (2015) estimated the PM_{2.5} concentration in China for 2006-2012 is 71 $\mu g \ m^{-3}$, higher than our work. According to the World Health Organization (WHO) Air Quality Interim Target-1, an annual mean PM_{2.5} concentration of 35 $\mu g \ m^{-3}$ or higher is associated with about 15% increased risk of premature mortality. As shown in Table 1, population-weighted PM_{2.5} for eastern China considerably exceeds
- the Interim Target-1 level of PM_{2.5} concentration, especially in Beijing and Shandong regions where the PM_{2.5} concentration is almost triple the Interim Target-1 level. These elevated concentrations threaten the health of <u>433</u> million inhabitants (Table 1) in eastern China who live in regions that exceed this target.

4. Conclusions

We estimated the ground-level concentration of PM_{2.5} in eastern China for 2013 using AOD retrieved from the GOCI satellite instrument, coupled with the relationship of AOD to PM_{2.5} simulated by a global chemical transport model (GEOS-Chem). GOCI-derived PM_{2.5} was compared with *in-situ* measurements throughout eastern China.

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JUNWEI XU 2015-8-16 5:04 PM Deleted: in situ JUNWEI XU 2015-8-16 5:04 PM Formatted: Font:Italic We applied a set of filters to GOCI AOD to remove cloud contamination. The filtered GOCI AOD showed significant agreement with AERONET AOD at Beijing and northern Taiwan (MFB of 6.7% to -1.2%). We also evaluated the simulated relationship of PM_{2.5} and AOD from GEOS-Chem by using an empirical relationship calculated from nearly

collocated ground-based PM2.5 monitors and AERONET AOD stations. A high degree of 5 consistency was observed between the GEOS-Chem simulation and ground-based measurements with MFB of -0.52% to 8.0%.

The GOCI-derived PM_{2.5} were highly consistent with *in-situ* measurements, capturing the similar seasonal and spatial distribution throughout eastern China. The highest PM2.5 concentrations were found in winter over northern regions. The annual averages of GOCI-derived PM_{2.5} were strongly correlated ($r^2=0.66$) with surface measurements with a slope near unity (1.01). Monthly comparison of GOCI-derived PM2.5 with ground-based measurements across the entire region of eastern China was also in good agreement with rRMSE =18.9%. The exclusion of our cloud-screening filters in GOCI retrievals would

15 introduce significant bias in GOCI-derived PM2.5, especially in summer and would increase the <u>rRMSE</u> by a factor of 1.7 - 5.3,

The chemical <u>composition</u> of GOCI-derived PM_{2.5} revealed that secondary inorganic aerosols (SIA; SO_4^{2-} , NO_3^{-} , NH_4^+) and organic matter (OM) dominated throughout the year. NO3- had a winter maximum due to aerosol thermodynamics. OM increased by a

20 factor of 2 in winter, which was primarily driven by biofuel emission for heating in northern China. Dust played an important role in spring and fall.

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The population-weighted GOCI-derived $PM_{2.5}$ for 2013 at 6 km by 6 km resolution in eastern China was 53.8 µg m⁻³, suggesting ~400 million people in China live in regions with $PM_{2.5}$ concentrations exceeding the suggested 35 µg m⁻³ by the World Health Organization (WHO) Air Quality Interim Target-1, of which ~130 million people in Beijing and Shandong regions are seriously threatened by even higher $PM_{2.5}$ concentrations. Population-weighted $PM_{2.5}$ of pixels containing ground-based monitors is

much higher at 82.4 μ g m⁻³, suggesting the value of the newly established PM_{2.5} network to monitor these seriously polluted regions.

The satellite measurements of AOD from the GOCI instrument coupled with the relationship between AOD and PM_{2.5} simulated by a chemical transport model have the potential to provide a unique synopsis of ground-level PM_{2.5} concentrations at fine spatial resolution in the most polluted and populated part of China. <u>Further development of this</u> capability will depend on both the quality of GOCI aerosol products and the aerosol simulation. Assimilating satellite observations of trace gases from the forthcoming GEMS (Geostationary Environment Spectrometer) geostationary platform would provide

additional constraints on PM2.5 composition.

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20 Sciences and Engineering Research Council of Canada) and by an Izaak Walton Killiam Memorial Scholarship for J. Xu. Computational facilities are partially provided by

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Canada.

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Figure 1. The GOCI granules from 5 July 2013, 10:30 (top) and 11:30 (bottom) Korean Standard Time. From left to right on each panel are the GOCI true color images, the operational AOD retrievals and the AOD retrievals after applying temporal and textual filters to reduce cloud contamination. The boxes highlight examples of challenging cloud fields, and are enlarged within the lower right subplot of each panel.



Figure 2. Top: Monthly time series of AOD from AERONET and GOCI for Jan - Dec 2013. Numbers above the x-axis denote the number of coincident hourly observations in each month. Bottom: Monthly averages of PM_{25} /AOD from ground measurements and the GEOS-Chem simulation at AERONET sites. The ground-based ratio is sampled from daily ground PM_{25} coincident with AERONET AOD for Jan - Dec 2013. The GEOS-Chem simulation is for May 2012 - April 2013, noncoincident with the ground-based ratio for May – Dec 2013. Numbers above the x-axis denote the number of daily ground-based observations in each month. Error bars represent standard errors <u>statistics</u> are root mean square error (RMSE), relative root mean square error (rRMSE), mean fractional bias (MFB) and mean fractional error (MFE).

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Figure 3. Seasonal and annual distribution of PM_{25} concentrations at 6 km by 6 km resolution over East Asia for 2013. The background color indicates averages of GOCI-derived daily surface PM_{25} concentrations. Filled circles represent averages of daily ground-based measurements of PM_{25} . Gray denotes missing values. Boxes in the annual map denote regions used for monthly comparisons in Fig. 5 from top to bottom: Beijing and surrounding areas, Shandong and surrounding regions, Shanghai and surrounding areas and northern Taiwan.

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Figure 4. Scatterplots of the annual mean (left) and seasonal mean (right) GOCI-derived PM₂₅ for 2013 against PM_{2.5} from 494 ground monitors over the GOCI domain in eastern China.

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Figure 5. Monthly averages of daily PM_{25} from *in-situ* measurements and daily PM_{25} estimated from GOCI AOD for 2013. Regions are defined in Fig. 3. Error bars represent standard errors. Statistics are root mean square error (RMSE), relative root mean square error (rRMSE), mean fractional bias (MFB) and mean fractional error (MFE),

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Figure 6. Monthly variation of GOCI-derived PM_{25} and <u>in-situ</u> PM_{25} for 2013 over eastern China, with chemical <u>composition</u> for GOCI-derived PM_{25} . The <u>in-situ</u> PM_{25} is determined from the averages of all ground stations in eastern China for 2013 and GOCI-derived PM_{25} is calculated from the average of all grid boxes that contain PM_{25} ground monitors. The chemical <u>composition</u> is calculated by applying the GEOS-Chem simulated mass fraction of $PM_{2.5}$ chemical components to GOCI-derived $PM_{2.5}$ mass concentration. Aerosol water is associated with each $PM_{2.5}$ component according to its hygroscopicity. Error bars represent standard errors.

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Table 1. Annual PM_{25} concentrations, <u>area-weighted concentrations of chemical</u> <u>composition</u> and <u>affected</u> population <u>of $PM_{2.5}$ in regions outlined in Fig. 3 and in overall eastern China (excluding northern Taiwan)</u> for 2013. Aerosol water is not associated with each $PM_{2.5}$ component for consistency with measurement protocols. $PM_{2.5}$ concentration is at 35 % relative humidity. IT1 refers to the WHO air quality interim target-1 of 35 µg m³.

Pagian	Beijing	Shandong	Shanghai	Eastern	Northern
Region				China	Taiwan
$\begin{array}{c} \mbox{Population-weighted GOCI-derived PM}_{2.5} \\ (\mu g/m^3) \end{array}$	90.8	89.1	56.9	53.8	18.9
Area-weighted GOCI-derived $PM_{2.5}$ ($\mu g/m^3$)	86.5	89.1	51.0	44.3	23.6
SO4 ²⁻ (µg/m ³)	12.8	14.0	9.2	13.1	5.1
NO_3^- (µg/m ³)	14.5	16.1	8.5	4.2	2.1
NH_4^+ (µg/m ³)	8.9	9.8	5.7	3.3	2.2
OC (µg∕m³)	10.3	9.6	4.3	2.9	1.6
BC (µg∕m³)	6.3	5.2	2.6	1.6	0.8
Dust (µg∕m³)	9.1	8.3	4.9	4.4	2.9
Sea Salt (µg∕m³)	0.2	0.4	0.9	1.9	2.2
OM (µg/m³)	17.1	15.7	7.4	5.4	3.0
Population (million people) exposed to PM _{2.5} exceeding IT-1 level	37.8	88.8	98.3	432.8	1.5

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