

1 **Supplement**

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3 **1. Comparison of simulated and OMI SO₂ and NO₂ columns**

4 We calculate the trends in simulated and OMI SO₂ and NO₂ columns (Figures S1 and S2)
5 to evaluate our GEOS-Chem simulation. There is broad consistency between the trends in our
6 simulated SO₂ and NO₂ columns with those from OMI. There are negative trends in both OMI and
7 simulated SO₂ columns over most of North America, Europe, northern South America, central
8 Africa, and east China. There is a mixture of negative and positive trends in SO₂ over North Africa.
9 There are positive trends in SO₂ over southern South America, southern Africa, the Middle-East,
10 India, most of China, and Australia. The trends in NO₂ columns correspond to the trends in SO₂
11 columns in almost all regions except for eastern China, which shows positive trends in NO₂
12 columns for both the simulation and OMI.

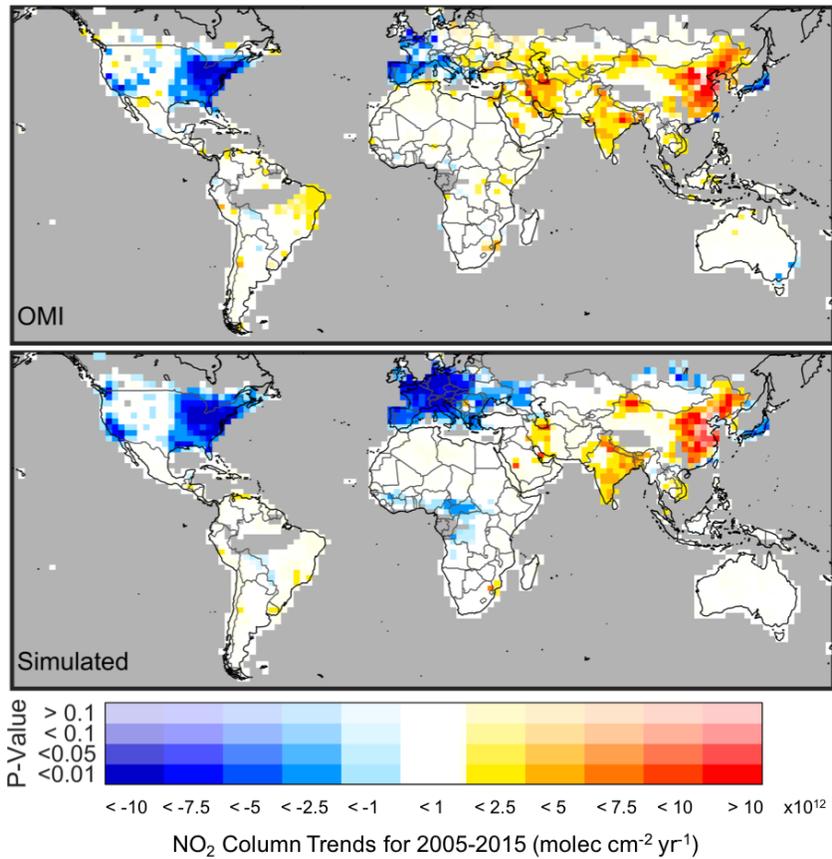
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14 **2. Comparison of simulated and satellite AOD**

15 Figure S3 shows the trends in GEOS-Chem and satellite AOD for 2005-2015 filtered based
16 on coincident OMI pixels with persistent cloud fraction greater than 5%. Overall the trends in
17 simulated AOD are consistent with the range of trends in satellite AOD. The GEOS-Chem AOD
18 (Figure S3a) shows negative trends in AOD over the eastern United States and West Africa, and
19 positive trends over the western United States, the Middle-East, India, and most of China. Figure
20 S3b shows the trends in AOD from MISR. Significant negative trends are apparent over the eastern
21 United States, Europe, central South America, parts of North Africa, West Africa, and
22 Mongolia/Inner Mongolia. There are small positive trends over west and central United States,
23 parts of South America, parts of North Africa, southern Africa, parts of the Middle-East, parts of
24 China, and Australia, with stronger positive trends over India. AOD from MODIS Dark Target
25 (Figure S3c) shows negative trends over eastern United States, Europe, and central South America,
26 with small positive trends over southern Africa, most of Asia, and Australia, and stronger positive
27 trends over Canada, southern South America, India, and over Central Asia between the Caspian
28 Sea and the Aral Sea. Figure S3d shows the trends in AOD from MODIS Deep Blue. There small
29 negative trends over eastern United States, central South America, Europe, parts of North Africa
30 and West Africa, with stronger negative trends over the Indo-Gangetic Plain and Mongolia/Inner
31 Mongolia. There are positive trends over southern Africa, most of Asia, and Australia, and stronger

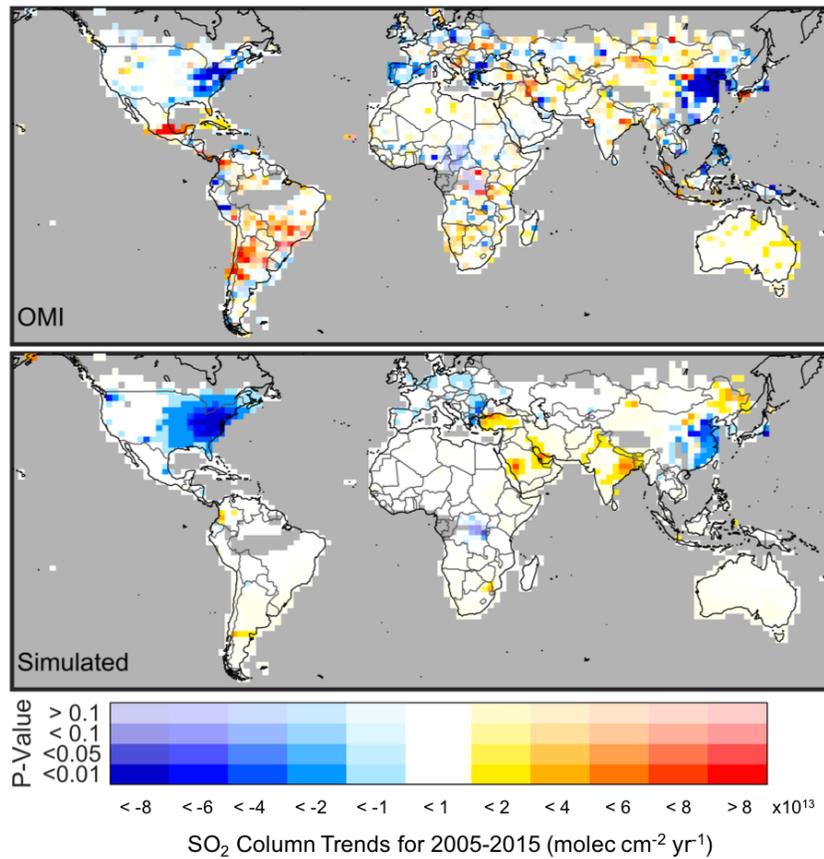
32 positive trends over Canada, southern South America, parts of the Middle-East, India, and over
33 Central Asia between the Caspian Sea and the Aral Sea. Figures S3e and S3f show the trends in
34 AOD from the OMI OMAERUV algorithm at 388 nm and 500 nm, respectively. Significant
35 negative trends are apparent for both wavelengths over central South America, West Africa, the
36 Indo-Gangetic Plain, and Mongolia/Inner Mongolia. Negative trends over Europe and parts of
37 North Africa are more pronounced in the OMI AOD at 388 nm (Figure S3e) than at 500 nm (Figure
38 S3f). There are small positive trends over west and central United States, parts of South America,
39 parts of North Africa, southern Africa, parts of China, and Australia, with stronger positive trends
40 over Canada, India, and over Central Asia between the Caspian Sea and the Aral Sea.

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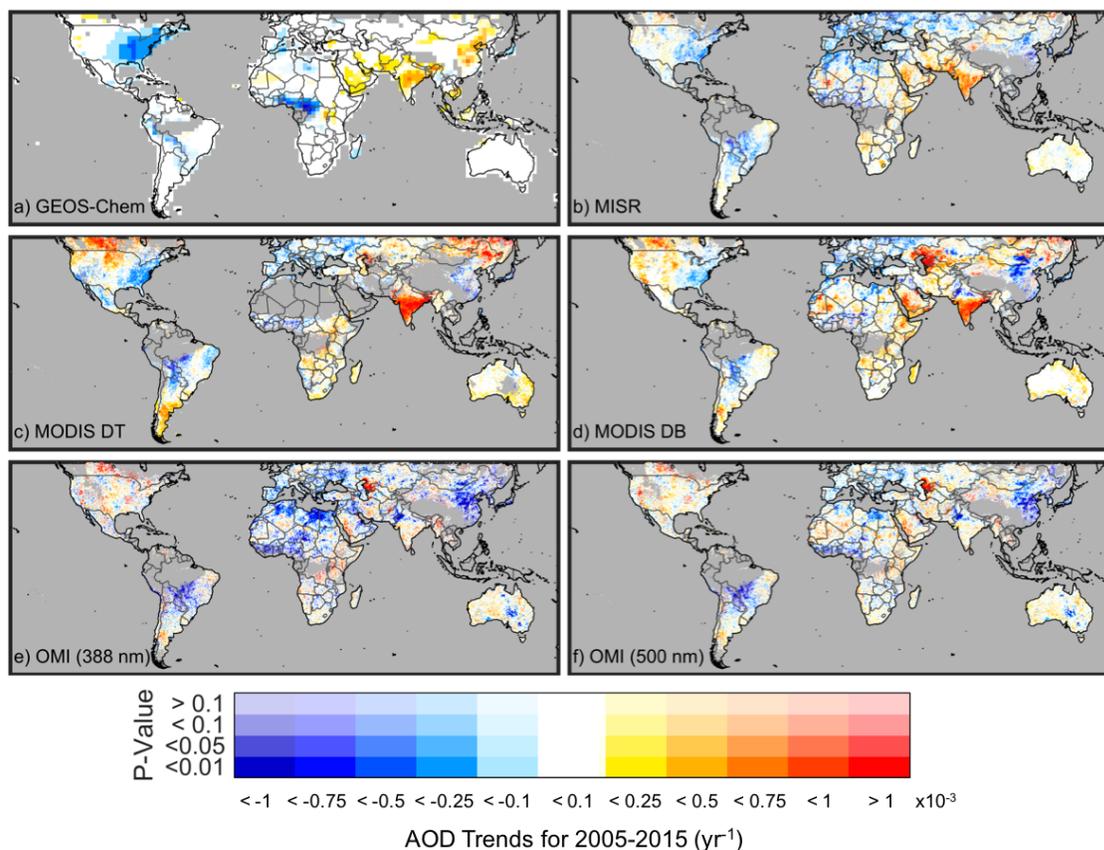
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48 **Figure S1:** Trends in OMI (top panel) and GEOS-Chem (bottom panel) NO₂ columns calculated
 49 from the Generalized Least Squares regression of monthly time series values over 2005-2015. The
 50 OMI NO₂ columns are from NASA's OMNO2 version 2.1 product. The opacity of the colors
 51 indicates the statistical significance of the trend. Gray indicates persistent cloud fraction greater
 52 than 5%.



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54 **Figure S2:** Trends in OMI (top panel) and GEOS-Chem (bottom panel) SO₂ columns calculated
 55 from the Generalized Least Squares regression of monthly time series values over 2005-2015. The
 56 OMI SO₂ columns are from NASA's OMSO₂ version 1.2.0 product. The opacity of the colors
 57 indicates the statistical significance of the trend. Gray indicates persistent cloud fraction greater
 58 than 5%.



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 60 **Figure S3:** Trends in aerosol optical depth from a) GEOS-Chem (550 nm), b) MISR v22 (550
 61 nm), c) MODIS Terra collection 6 Dark Target algorithm (550 nm), d) the MODIS Terra collection
 62 6 Deep Blue algorithm (550 nm), and the OMI OMAERUV algorithm for e) 388 nm and f) 500
 63 nm. The GEOS-Chem simulation is sampled coincidentally with the OMI UVAI product. The trends
 64 are calculated from the Generalized Least Squares regression of monthly time series values over
 65 2005-2015. The opacity of the colors indicates the statistical significance of the trend. Gray
 66 indicates persistent cloud fraction greater than 5%.