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## Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States

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[1] We use satellite (MODIS) observations of aerosol optical depths (AODs) over the North Pacific, together with surface aerosol measurements at a network of remote U.S. sites (IMPROVE), to improve understanding of the transpacific transport of Asian aerosol pollution and assess the ability of a global 3-D chemical transport model (GEOS-Chem CTM) to quantify Asian aerosol enhancements in U.S. surface air. The MODIS observations show the strongest transpacific transport occurring in spring at 40–55°N. This transport in the model takes place mainly in the lower free troposphere (900–700 hPa) because of scavenging during transport either in the boundary layer or during lifting to the upper troposphere. The preferential altitude of aerosol transpacific transport results in direct impact on the elevated terrain of the NW United States. Sulfate observations in the NW United States in spring 2001 show higher concentrations on the days of model-predicted maximum Asian influence ( $1.04 \mu\text{g m}^{-3}$ ) than seasonal mean values ( $0.69 \mu\text{g m}^{-3}$ ). No such Asian enhancements are observed for nitrate or for organic carbon (OC) aerosol. Distinct Asian sulfate episodes correlated with dust events are observed in the NW United States and simulated with the model. The mean Asian pollution enhancement in that region in spring is  $0.16 \mu\text{g m}^{-3}$  with a 50% estimated uncertainty. This is higher than the estimated natural concentration of  $0.09 \mu\text{g m}^{-3}$  presently used as objective for regulation of visibility in U.S. wilderness areas.

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### 1. Introduction

[2] Episodes of elevated sulfate aerosol concentrations (up to  $1.8 \mu\text{g m}^{-3}$ ) in air masses of Asian origin have been observed at sites in the northwestern United States [Jaffe *et al.*, 1999, 2001, 2003a; Bertschi *et al.*, 2004]. Simulations using the GEOS-Chem global 3-D chemical transport model (CTM) indicate an annual mean sulfate enhancement of  $0.10 \mu\text{g m}^{-3}$  in surface air over the western United States due to Asian anthropogenic emissions [Park *et al.*, 2006]. Although such enhancements are small relative to the  $15 \mu\text{g m}^{-3}$  annual air quality standard for fine aerosol in the United States [U.S. Environmental Protection Agency (U.S. EPA), 1996], they degrade visibility significantly

relative to the “natural visibility conditions” set as a target by the EPA Regional Haze Rule for U.S. national parks [U.S. EPA, 2003] and have important implications for present application of the Rule [Park *et al.*, 2004]. There is a need to better understand the transpacific transport of anthropogenic Asian aerosols and the ability of models such as GEOS-Chem to quantify it. We address this issue here through a concerted analysis of satellite and ground-based aerosol observations.

[3] Transport of Asian anthropogenic aerosols across the Pacific was first documented in the 1980s from observations at island sites [Prospero *et al.*, 1985, 2003; Ziemann *et al.*, 1995; Arimoto *et al.*, 1996; Huebert *et al.*, 2001]. These revealed a spring maximum in transpacific transport, coinciding with the seasonal maximum in Asian dust emission, and resulting in combined dust and pollution signatures. Aircraft observations in Asian outflow over the NW Pacific [Arimoto *et al.*, 1997; Jordan *et al.*, 2003; Maxwell-Meier *et al.*, 2004], and in transpacific Asian plumes over the NE Pacific [Andreae *et al.*, 1988; Clarke *et al.*, 2001; Price *et al.*, 2003] provided subsequent evidence of aerosol transport in the lower free troposphere, including coincident sulfate and dust. More recently, satellite retrievals of aerosol optical depth (AOD) have been used to track Asian dust and pollution plumes across the Pacific [Husar *et al.*, 1997,

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2001; *Edwards et al.*, 2004; *Darmenova et al.*, 2005]. A model study by *Takemura et al.* [2002] examined the signatures of Asian dust storms over the Pacific and found that the contribution of anthropogenic aerosol to the total AOD is comparable to that of the dust.

[4] Our objective here is to determine the combined constraints from the space-based and surface-based observations for better understanding the mechanisms involved in aerosol transpacific transport and for quantifying Asian anthropogenic influence on surface aerosol concentrations in the United States. We apply the GEOS-Chem CTM to the interpretation of AOD observations over the Pacific from the MODIS satellite instrument and from the AERONET surface-based network, together with aerosol observations in surface air over the United States from the IMPROVE network. We focus on the year 2001, which has been used in previous GEOS-Chem analyses interpreting aircraft observations of Asian outflow and transpacific transport from the TRACE-P aircraft campaign [*Liu et al.*, 2003; *Palmer et al.*, 2003; *Heald et al.*, 2003a, 2003b, 2004; *Park et al.*, 2005].

## 2. Model Simulation

[5] We conduct a global coupled oxidant-aerosol simulation for 2001 using the GEOS-Chem CTM v7.01.02 (<http://www-as.harvard.edu/chemistry/trop/geos/index.html>). This simulation is driven by assimilated meteorological data with  $1^\circ \times 1^\circ$  horizontal resolution, 48 vertical layers, and a temporal resolution of 6 hours (3 hours for surface variables and mixing depths) from the Goddard Earth Observing System (GEOS)-3 of the NASA Global Modeling and Assimilation Office (GMAO). The horizontal resolution of the meteorological data is degraded to  $2^\circ \times 2.5^\circ$  for input to GEOS-Chem.

[6] The GEOS-Chem oxidant-aerosol simulation includes  $\text{H}_2\text{SO}_4$ - $\text{HNO}_3$ - $\text{NH}_3$  aerosol thermodynamics coupled to an ozone- $\text{NO}_x$ -hydrocarbon-aerosol chemical mechanism [*Park et al.*, 2004]. It also includes organic carbon (OC) and elemental carbon (EC) aerosols [*Park et al.*, 2003], sea salt aerosol [*Alexander et al.*, 2005], and soil dust [*Fairlie et al.*, 2004]. Secondary organic aerosol (SOA) is produced from the oxidation of biogenic hydrocarbons following the *Chung and Seinfeld* [2002] scheme. The aerosol and oxidant chemistry are coupled through aerosol formation (sulfate, nitrate, SOA), heterogeneous reactions, and aerosol effects on photolysis rates [*Martin et al.*, 2003]. GEOS-Chem simulates aerosol mass concentrations, and does not include aerosol microphysics.

[7] Optical properties are calculated in GEOS-Chem for each aerosol component as a function of local relative humidity (RH), as described by *Martin et al.* [2003]. All aerosols are treated as externally mixed with lognormal size distributions and optical properties defined by the Global Aerosol Data Set (GADS) [*Köpke et al.*, 1997] database and by *Ginoux et al.* [2001] and *Patterson et al.* [1977] in the case of dust. The AOD at 550 nm is calculated within GEOS-CHEM for local conditions from the mass concentration, extinction efficiency, effective radius, and particle mass density according to the formulation of *Tegen and Lacis* [1996]. The organic aerosol mass used in optical calculations is taken to be twice the OC mass tracked in the

model, to account for the noncarbon component of organic mass following *Turpin and Lim* [2001].

[8] All aerosols are subject to dry deposition and hydrophilic aerosols are subject to wet deposition. Wet deposition includes rainout and washout from large-scale precipitation as well as scavenging in convective updrafts [*Liu et al.*, 2001]. Conversion of hydrophobic to hydrophilic carbonaceous aerosols takes place with an e-folding time of 1.2 days based on *Cooke et al.* [1999]. The scheme provides a good simulation of soluble species concentrations in the United States [*Park et al.*, 2003; 2004] and in North American and Asian outflow [*Li et al.*, 2005; *Park et al.*, 2005]. Dry deposition of dust and sea-salt aerosols follows the size-dependent scheme of *Zhang et al.* [2001] and accounts for hygroscopic growth as a function of relative humidity. Dry deposition for all other aerosols is simulated with a standard resistance-in-series scheme based on *Wesely* [1989], as described by *Wang et al.* [1998].

[9] The global sources of sulfur, ammonia, and  $\text{NO}_x$  are as described by *Park et al.* [2004]. Carbonaceous aerosol emissions are taken from *Cooke et al.* [1999] for fossil fuel, *Yevich and Logan* [2003] for biofuels, and *Park et al.* [2006] for biomass burning. Asian emissions of EC aerosols, OC aerosols, and sulfur oxides total  $4.2 \text{ TgC yr}^{-1}$ ,  $16 \text{ TgC yr}^{-1}$ , and  $24 \text{ TgS yr}^{-1}$ , respectively. Sea salt and dust emission follow the schemes of *Monahan et al.* [1986] and *Zender et al.* [2003]. Several major Asian dust events took place during April 2001 [*Takemura et al.*, 2002; *Thulasiraman et al.*, 2002; *Jaffe et al.*, 2003b] and are captured by the model [*Fairlie et al.*, 2004]. Globally, 1380 Tg of dust are emitted in the model in 2001, a fifth of which comes from Asia, where the dust source is particularly strong during the spring season.

[10] Observations in the marine boundary layer (MBL) indicate a marine source of OC aerosol, which appears to be of primary biological origin and follows the same bubble-bursting mechanism that generates sea salt aerosols [*Matthias-Maser et al.*, 1999; *Putaud et al.*, 2000; *Kleefeld et al.*, 2002; *O'Dowd et al.*, 2004]. To our knowledge this source has not been included in previous CTMs. Our initial simulation with no marine source yielded OC concentrations of less than  $0.05 \mu\text{g m}^{-3}$  in the MBL, whereas observations are in the  $0.05\text{--}1 \mu\text{g m}^{-3}$  range [*Penner*, 1995, and references therein; *Novakov et al.*, 1997; *Putaud et al.*, 2000; *Kleefeld et al.*, 2002; *Neusüß et al.*, 2002; *O'Dowd et al.*, 2004; *Jaffe et al.*, 2005]. We thus introduced in GEOS-Chem a marine OC aerosol source by convolving the sea salt aerosol source with the seasonal variation of marine primary productivity observed by MODIS [*Savtchenko et al.*, 2004], and scaling globally this source to match mean OC aerosol measurements over the remote oceans. We assume that marine OC is emitted in the same size distribution as sea salt and is thus subject to rapid dry deposition [*Alexander et al.*, 2005]. The resulting global marine OC emission in the model is  $27 \text{ Tg C yr}^{-1}$ , with simulated concentrations of  $0.04\text{--}0.4 \mu\text{g m}^{-3}$  in the remote MBL. This source amounts to about half of the global continental OC source, but the short lifetime of the coarse sea salt particles (averaging 8 hours) confines its importance to the MBL.

[11] The GEOS-Chem aerosol simulation has been evaluated in a number of previous studies. GEOS-Chem gives a

generally unbiased simulation of sulfate and carbonaceous aerosol concentrations over North America [Park *et al.*, 2003, 2004]. Park *et al.* [2005] showed that the model can reproduce with no significant bias the observed vertical profiles of sulfate, nitrate, and EC aerosols from the TRACE-P aircraft campaign, and the corresponding export efficiencies as determined from correlation with CO. This is particularly important in supporting the ability of the model to quantify transpacific Asian influence. The Asian EC source bias pointed out by Park *et al.* [2005] has been corrected here with the use of the Cooke *et al.* [1999] emission inventory. High OC aerosol concentrations ( $1\text{--}4\ \mu\text{g m}^{-3}$  STP) observed from aircraft up to 7 km altitude off the coast of Japan [Maria *et al.*, 2003] are considerably underestimated by GEOS-Chem and imply a missing SOA source in the free troposphere [Heald *et al.*, 2005]. We have not attempted to account for this source in the work presented here. Global model treatments of aerosols and simulated AOD are reviewed and evaluated by Kinne *et al.* [2003]. The GEOS-Chem aerosol simulation is similar to the GOCART model of Chin *et al.* [2002] with modifications to sources and oxidation schemes as noted by Park *et al.* [2003, 2004].

[12] We conducted three simulations for 2001: a standard simulation as described above, and two sensitivity simulations (1) excluding Asian anthropogenic emissions and (2) excluding Asian natural emissions (dust, biogenic, and biomass burning). The definitions of anthropogenic and natural follow the earlier work of Park *et al.* [2003, 2004]. From these simulations we quantify the contribution of Asian sources to transpacific enhancements and surface concentrations over the United States.

### 3. MODIS AODs as Indicators of Transpacific Transport

[13] MODIS observations on the NASA EOS-Terra satellite provide continuous data for AODs over the North Pacific in 2001 with near-complete spatial coverage every day, limited by cloud cover. The data are taken from a sun-synchronous polar orbit with 1030 local time overpass. We use the daily gridded AOD (550 nm) v4 product with  $1^\circ \times 1^\circ$  resolution [Chu *et al.*, 2002; Remer *et al.*, 2002] and regrid these observations to  $2^\circ \times 2.5^\circ$  for comparison to GEOS-Chem 550 nm AODs. Aerosol optical properties assumed in the model may be different from those assumed in the MODIS retrieval but we do not attempt to take into account this difference. MODIS retrieval of size parameters is not available for our period because of calibration errors [Chu *et al.*, 2005].

[14] Figure 1 shows the seasonal variation of AODs across the Pacific as seen by MODIS and simulated by GEOS-Chem. MODIS and GEOS-Chem show consistent seasonal variations with transpacific transport occurring primarily north of  $40^\circ\text{N}$  and peaking in spring. The spring maximum is driven in part by seasonal dust emission [Uematsu *et al.*, 1983] but also by frequent warm conveyor belts (WCBs) lifting East Asian anthropogenic outflow to the free troposphere where it is rapidly transported across the Pacific by strong westerlies [Liu *et al.*, 2003]. 2001 was an unusually high year for Asian dust [Takemura *et al.*, 2002; Thulasiraman *et al.*, 2002; Jaffe *et al.*, 2003b], yet we

find in the model that dust and pollution make contributions of similar magnitude to the springtime AOD enhancements over the Pacific, consistent with the work of Takemura *et al.* [2002]. The transpacific Asian pollution component in the model is mainly sulfate [Park *et al.*, 2004], because of relatively efficient escape of  $\text{SO}_2$  from the continental boundary layer followed by production during transport across the Pacific [Brock *et al.*, 2004]. We will examine in section 6 some specific springtime events of transpacific anthropogenic aerosol transport as seen by MODIS.

[15] The GEOS-Chem simulation is lower than MODIS throughout the year. Comparison over the North Pacific in spring, for the region in Figure 1 when the model is sampled for the MODIS overpasses, shows a strong spatial correlation ( $R = 0.88$ ), but the model AOD is a factor of 2 lower. Early validation papers comparing MODIS with column AOD observations from the AERONET network surface sites [Holben *et al.*, 1998] reported MODIS errors of  $\Delta\text{AOD} = \pm 0.05 \pm 0.2\text{AOD}$  over land and  $\Delta\text{AOD} = \pm 0.05 \pm 0.05\text{AOD}$  over ocean, with no significant bias [Chu *et al.*, 2002; Remer *et al.*, 2002]. However, more recent studies have found that the MODIS AOD product over land is biased high [Kinne *et al.*, 2003; Chin *et al.*, 2004; Matsui *et al.*, 2004; Hauser *et al.*, 2005]. Comparisons between MODIS and the GOCART CTM during the ACE-Asia period in April 2001 found MODIS to be a factor of 2–3 higher over Asia and North America [Chin *et al.*, 2004]. Nonsphericity effects associated primarily with dust have been shown to lead to MODIS overestimates of AODs over the Pacific [Chu *et al.*, 2005]. There is in addition an apparent bias under clean conditions at remote AERONET sites where MODIS AOD values are rarely below 0.1 [Kinne *et al.*, 2003; Eck *et al.*, 2005]. Cloud contamination is a further concern in the MODIS retrieval [Martins *et al.*, 2002; Zhang *et al.*, 2005], though we find no correlation between the MODIS cloud fraction and the model versus MODIS discrepancy.

[16] Figure 2 compares the simulated and MODIS AOD with observations from AERONET surface sites in the North Pacific. AERONET AOD measurements at the wavelengths used here have a well-calibrated uncertainty of  $\pm 0.01$  [Holben *et al.*, 1998]. Midway Island ( $28^\circ\text{N}$ ,  $177^\circ\text{W}$ ) experiences strong Asian influence [Prospero *et al.*, 2003]. MODIS captures the variability ( $R^2 = 0.68$ ) of the AERONET observations at this site and is high by 28% in the mean. GEOS-Chem underestimates the AERONET AODs at Midway by 33% on average; the corresponding  $R^2$  is only 0.41 but the model reproduces the timing of Asian transport events. We will discuss later four of these events (indicated by arrows). Part of the model underestimate at Midway could reflect the missing OC aerosol source in the free troposphere previously noted by Heald *et al.* [2005] in comparison with ACE-Asia observations of Asian outflow. The corresponding AOD underestimate would be 0.06 if the GADS OC size distribution and optical properties are assumed [Heald *et al.*, 2005]. At the Lanai (Hawaii) site at ( $21^\circ\text{N}$ ,  $157^\circ\text{W}$ ), and at the nearby Coconut Island site (not shown), MODIS shows large overestimates (over a factor of 2) and spurious variability while GEOS-Chem is in better agreement with AERONET, exhibiting little variability and a mean underestimate of 5–24%. Hawaii is outside of the main Asian



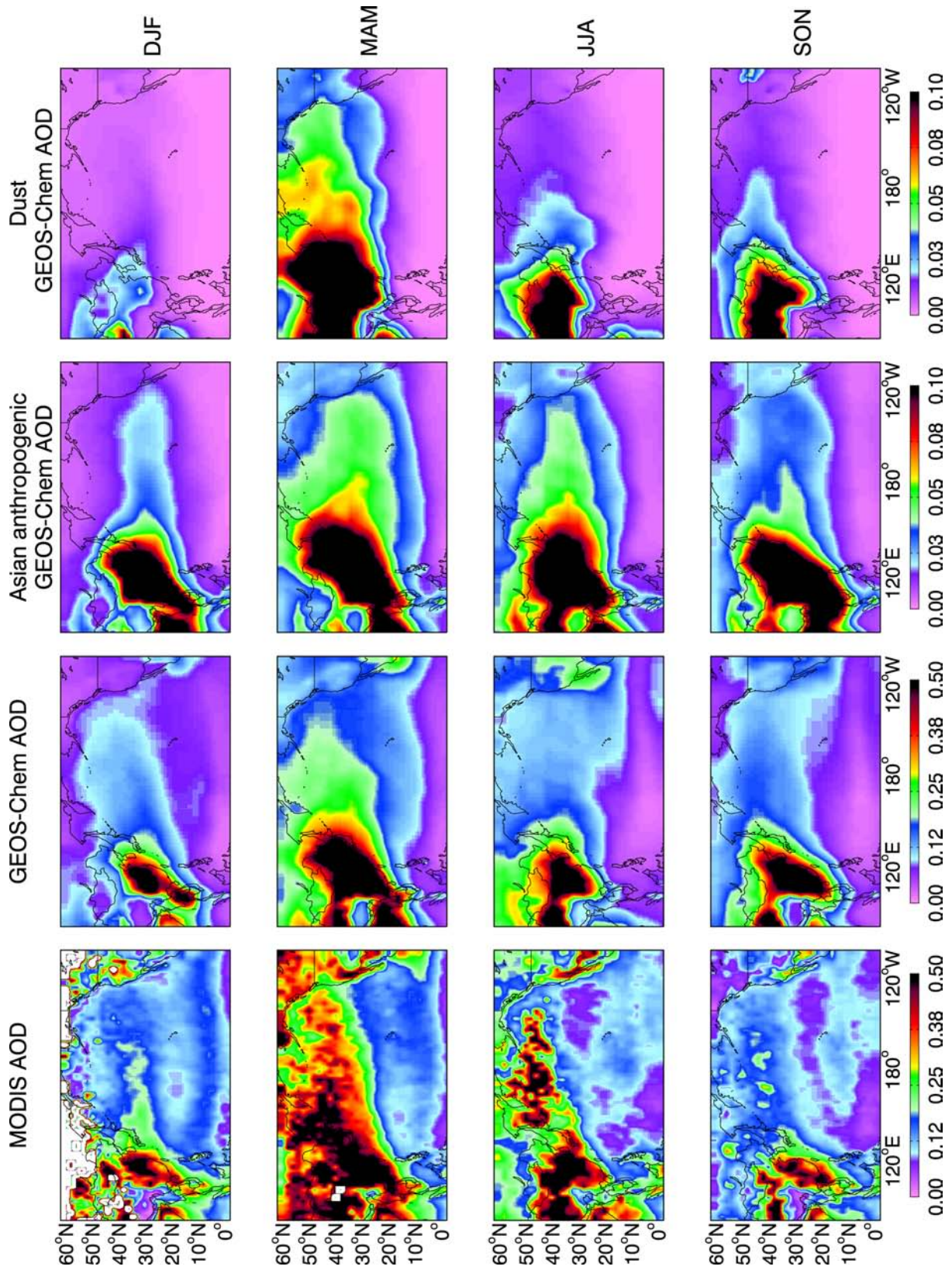
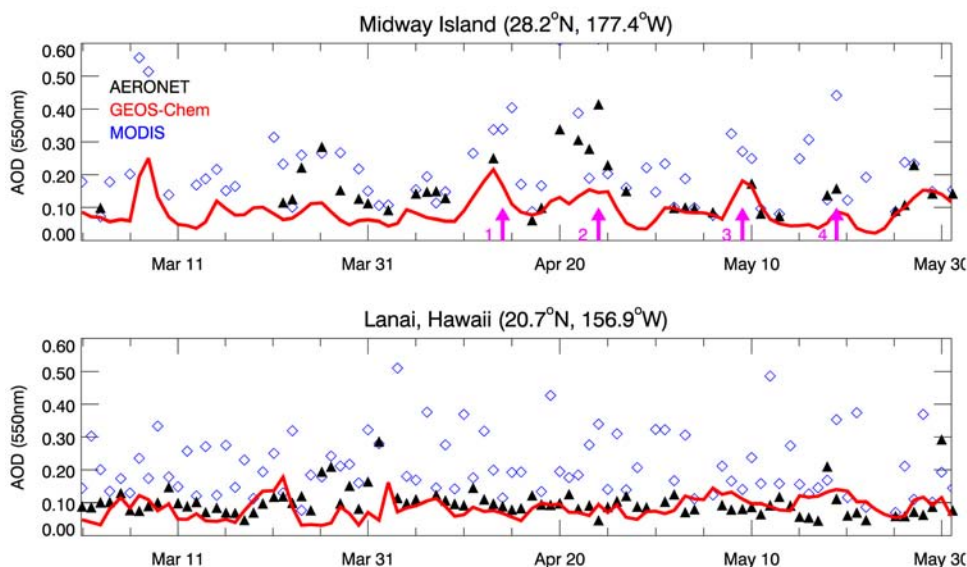


Figure 1



**Figure 2.** AOD time series at North Pacific AERONET sites in spring 2001 comparing data from AERONET (black triangles), MODIS (blue diamonds) and GEOS-Chem (red line). AERONET level 2 daily mean AOD observations in the 500 nm and 670 nm channels are interpolated to 550 nm for comparison with MODIS. Transpacific transport events at Midway discussed later in the text are highlighted with purple arrows.

aerosol transport route as seen by both the model and MODIS (Figure 1).

#### 4. Differential Transpacific Transport of Aerosols and CO

[17] Previous studies of transpacific pollution have focused on CO as a tracer of transport because of its long lifetime and the availability of observations from both aircraft and space [Yienger *et al.*, 2000; Heald *et al.*, 2003b; Jaeglé *et al.*, 2003; Edwards *et al.*, 2004; Hudman *et al.*, 2004; Liang *et al.*, 2004; Liu *et al.*, 2005]. Despite common combustion sources, we find that aerosol and CO transport across the Pacific differ both in timing and in spatial extent.

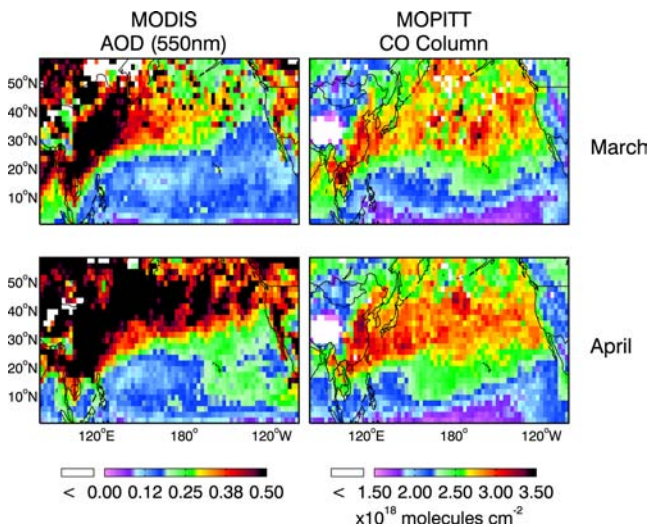
[18] As shown in Figure 3, the pattern of transpacific transport of CO observed by the MOPITT satellite instrument [Deeter *et al.*, 2003], also aboard the EOS-Terra platform, extends further south than the MODIS aerosol. Biomass burning in Southeast Asia is a major component of the springtime Asian outflow for CO [Chan *et al.*, 2003; Heald *et al.*, 2003a]. Burning extends from northeast India to southern China, and is strongest in Burma and Thailand (Figure 3). Biomass burning effluents are transported over the Pacific at lower latitudes than Asian anthropogenic pollution [Heald *et al.*, 2003b]. The MODIS AOD observations in spring indicate little transpacific transport of

biomass burning aerosols emitted from Southeast Asia. Although biomass burning is a major source of carbonaceous aerosol, it appears that these aerosols are efficiently scavenged during convective lifting over SE Asia. We show both March and April in Figure 3 to demonstrate that there is a latitudinal distinction in transport between aerosols and CO even in March when the Asian dust source was minimal.

[19] Figure 4 shows the longitude-altitude cross sections of simulated biomass burning Asian CO, anthropogenic Asian CO, and anthropogenic Asian aerosols across the North Pacific at 25–55°N in spring 2001. Transpacific transport of CO takes place at all altitudes, with dominant contribution from anthropogenic pollution in the lower to middle troposphere, and from biomass burning in the middle to upper troposphere driven by deep convective lifting [Liu *et al.*, 2003]. By contrast, transpacific transport of anthropogenic aerosols is narrowly focused in the lower free troposphere (900–700 hPa). The major distinction between the simulated transpacific transport of anthropogenic aerosol and CO is the lack of aerosol transport in the boundary layer, previously found by Liang *et al.* [2004] to be important for CO. Although strong Asian aerosol outflow was observed in the MBL as well as in the lower free troposphere during the TRACE-P and ACE-Asia aircraft missions [Browell *et al.*, 2003; Bates *et al.*, 2004; Merrill and Kim, 2004], we see from Figure 4 that the MBL

**Figure 1.** Seasonal variation of aerosol optical depth (AOD) at 550 nm over the North Pacific in 2001. Mean gridded values ( $2^\circ \times 2.5^\circ$ ) are shown for MODIS, GEOS-Chem, and the Asian anthropogenic and dust components of GEOS-Chem. The Asian anthropogenic component of the aerosol in GEOS-Chem is determined by difference between the standard simulation and a sensitivity simulation with Asian anthropogenic emissions shut off. Biomass burning is not included as anthropogenic. Color scales are saturated at the maximum values indicated in the legend. Model AODs are seasonal means.





**Figure 3.** Transpacific transport of aerosols and CO during March and April 2001. Shown are monthly mean AODs and CO columns observed by MODIS and MOPITT. We use daily MOPITT column version 3 data (<http://www.eos.ucar.edu/mopitt/>). Color scales are saturated at maximum values.

transport does not extend beyond the central Pacific because of deposition losses. Asian aerosol transported across the Pacific in the lower free troposphere may directly affect surface air in the western United States because of the elevated terrain (Figure 4).

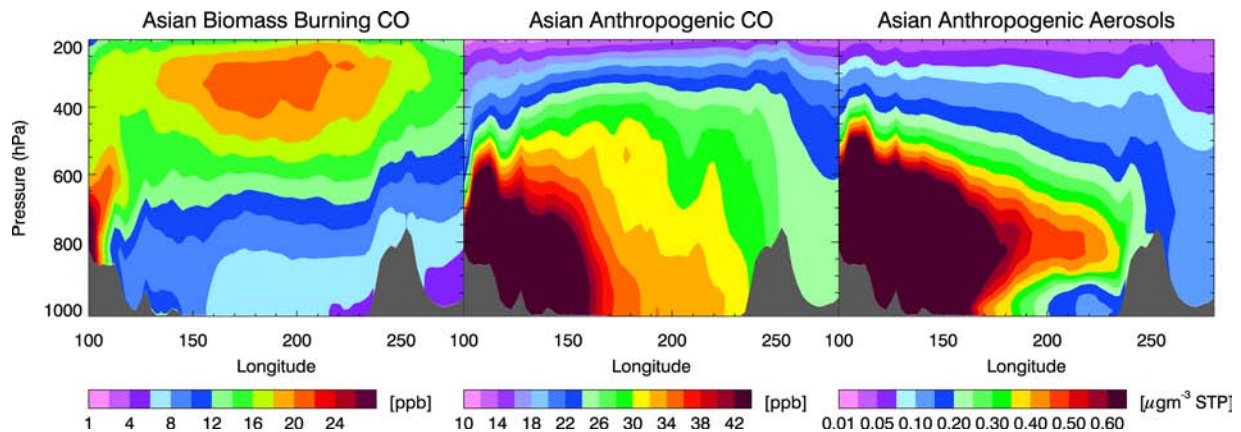
### 5. Asian Sulfate Enhancements at U.S. Surface Sites

[20] We now present evidence for Asian pollution influence on surface aerosol concentrations in the United States, using IMPROVE observations for the spring of 2001, and derive quantitative constraints for the magnitude of this

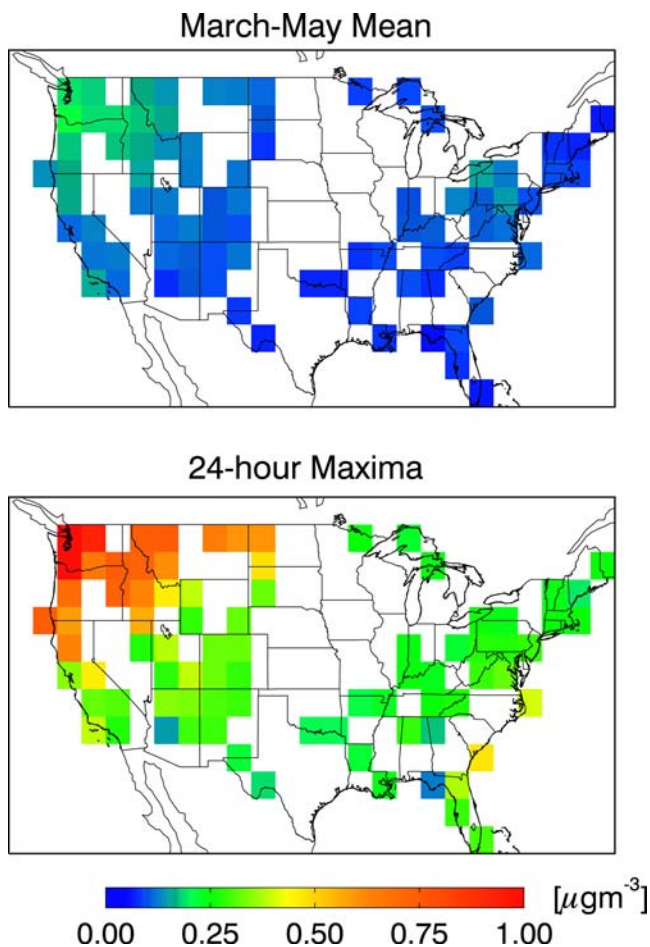
influence. The Interagency Monitoring for Protected Visual Environments (IMPROVE) network of stations was established in 1987 to monitor visibility in national parks in the United States [Malm *et al.*, 1994]. Surface concentrations of sulfate, nitrate, OC, and EC are measured as 24-hour averages every third day. We use observations from the 126 active IMPROVE sites in the continental United States during spring 2001. Park *et al.* [2004] previously showed comparisons of simulated and observed annual mean concentrations at these sites for 2001. We focus on sulfate as it is the main component of transpacific anthropogenic aerosol, at least in the model [Park *et al.*, 2004], and also on dust as an Asian tracer. We sample the GEOS-Chem model as 24-hour averages on the IMPROVE observation days in order to compare to the IMPROVE data.

[21] Figure 5 shows the springtime mean and 24-hour maximum Asian enhancements of sulfate concentrations in surface air simulated by the model at the IMPROVE sites. Asian influence is strongest in the northwest, consistent with the latitudinal structure of transpacific transport seen by MODIS (Figure 3). Maximum influence is in Washington state and southern British Columbia. For subsequent analysis we focus on the NW United States region extending west of 100°W and north of 40°N. The simulated springtime mean Asian enhancement of sulfate concentrations in that region is  $0.16 \mu\text{g m}^{-3}$  (Figure 5), as compared to  $0.10 \mu\text{g m}^{-3}$  on an annual mean basis [Park *et al.*, 2006]. The simulated 24-hour maximum enhancements average  $0.60 \mu\text{g m}^{-3}$  for the region.

[22] We searched for evidence of Asian influence in the IMPROVE data by comparing the observed seasonal mean concentrations to those observed on the days when the model diagnoses maximum Asian influence. Results are shown in Figure 6. The mean observed sulfate in the NW United States on the days of simulated maximum Asian influence ( $1.04 \mu\text{g m}^{-3}$ ) is indeed higher than the observed seasonal mean ( $0.69 \mu\text{g m}^{-3}$ ). The difference of  $0.35 \mu\text{g m}^{-3}$  is similar to the corresponding difference in the model ( $0.44 \mu\text{g m}^{-3}$ ) and provides support for the magnitude of



**Figure 4.** Simulated cross sections of Asian biomass burning CO, anthropogenic CO, and anthropogenic aerosol mixing ratios across the North Pacific at 25–55°N latitude during March–May 2001, as a function of longitude and altitude. Asian influences are determined by difference of the standard simulation with sensitivity simulations shutting off the corresponding sources. Topography at 40°N is shaded in grey. STP refers to standard temperature and pressure (273 K, 1013 hPa). Color scales are saturated at maximum values.



**Figure 5.** Asian anthropogenic enhancements of sulfate concentrations in surface air during the spring of 2001 (March–May) as simulated by the GEOS-Chem model. Values are shown for model grid squares corresponding to the locations of the IMPROVE network sites. The Asian anthropogenic component of the aerosol in GEOS-Chem is determined by difference between the standard simulation and a sensitivity simulation with Asian anthropogenic emissions shut off. Seasonal means and 24-hour maxima are shown. The color scale is saturated at  $1 \mu\text{g m}^{-3}$ .

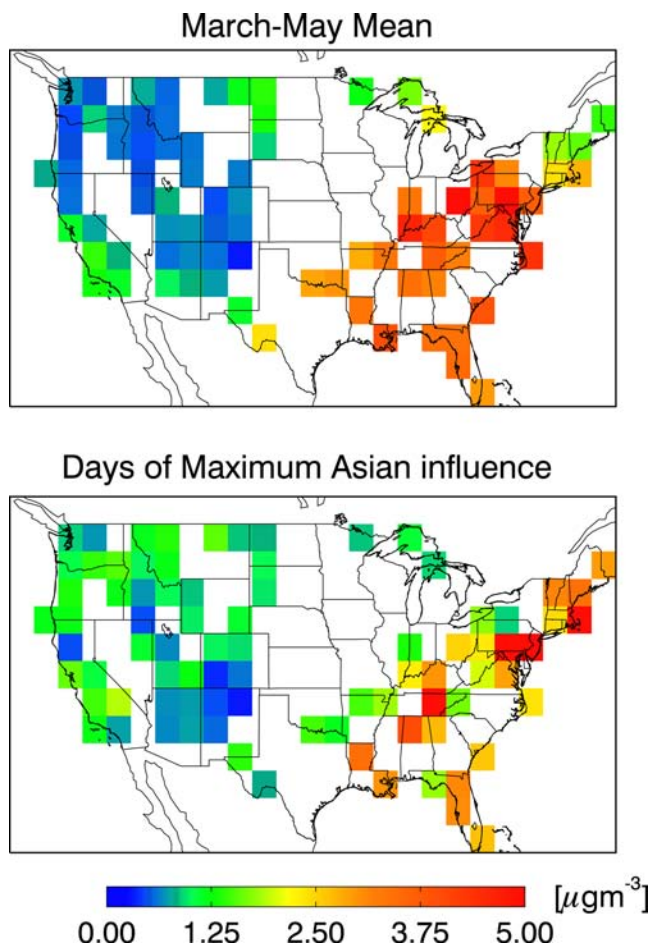
Asian influence as simulated by the model. In the eastern United States, by contrast, the days of maximum Asian influence have observed sulfate concentrations lower than the seasonal mean (Figure 6), reflecting the dominance of regional sources.

[23] The GEOS-Chem model predicts no significant Asian enhancements of nitrate and OC aerosols in the United States because of scavenging during Asian outflow. To test this result, we repeated the analysis of Figure 6 for observed nitrate and OC aerosol at the NW IMPROVE sites. We find that the regional springtime mean concentrations observed for nitrate ( $0.22 \mu\text{g m}^{-3}$ ) and OC ( $1.08 \mu\text{g m}^{-3}$ ) are not significantly different from those on the days of maximum Asian sulfate influence as simulated by GEOS-Chem ( $0.24 \mu\text{g m}^{-3}$  for nitrate and  $1.19 \mu\text{g m}^{-3}$  for OC).

[24] We examined the time series of observed and simulated concentrations at the NW IMPROVE sites for further

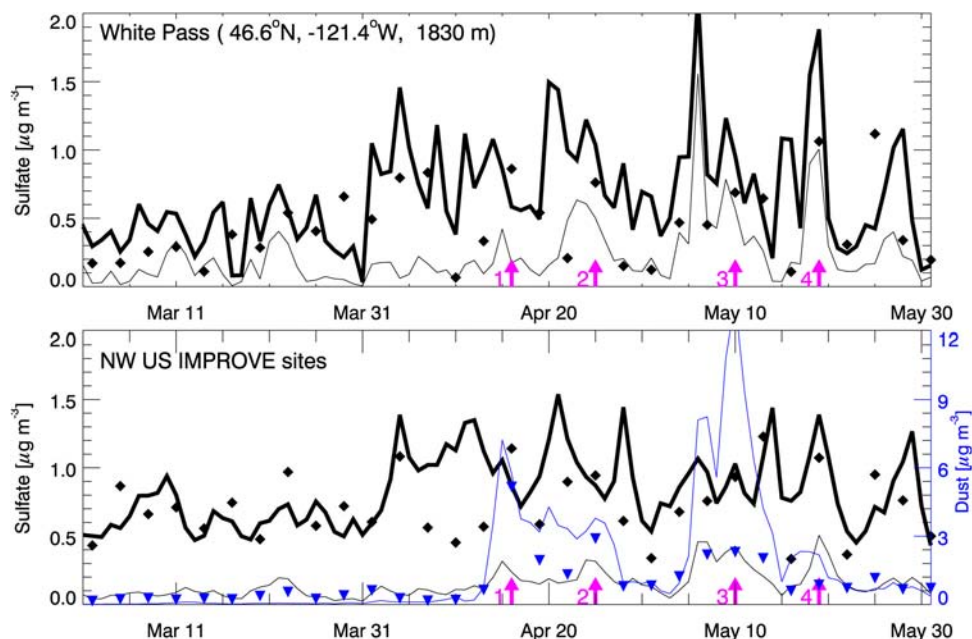
characterization of Asian influence. Figure 7 shows the simulated and observed time series of sulfate concentrations at White Pass, Washington ( $46^\circ\text{N}$ ,  $121^\circ\text{W}$ , 1.8 km altitude), a remote site where Asian influence is among the highest in the model (up to  $1.5 \mu\text{g m}^{-3}$  on a 24-hour maximum basis). Also shown are the time series averaged over the ensemble of NW sites. The model time series indicates several distinct pollution episodes. Autocorrelation analysis indicates a typical 4-day duration for the episodes, suggesting that the 3-day measurement frequency in the IMPROVE protocol should allow their detection. There is high spatial correlation of Asian pollution influence in the model across the ensemble of NW sites (simulated Asian aerosol concentrations are correlated at the 95% confidence level between sites that are separated by less than 1600 km), with no significant time lag, indicating that Asian events are regional in scale and supporting the use of spatial averaging in the time series.

[25] Also shown in Figure 7 are the simulated and observed time series of PM<sub>2.5</sub> dust concentrations at the ensemble of NW IMPROVE sites. High dust concentrations of Asian origin are found in both the observations and the model for the period extending from 10 April to 31 May.



**Figure 6.** Observed sulfate concentrations at IMPROVE sites during the spring of 2001 (March–May): seasonal means and values sampled on the days of the 24-hour local model maxima in Asian influence (see Figure 5). The color scale is saturated at  $5 \mu\text{g m}^{-3}$ .





**Figure 7.** Time series of sulfate concentrations in surface air (top) at White Pass, Washington, and (bottom) for the ensemble of IMPROVE sites in the NW United States (west of  $100^{\circ}\text{W}$  and north of  $40^{\circ}\text{N}$ ). The IMPROVE observations are shown as diamonds, and the GEOS-Chem model values are shown as thick solid lines; correlation between the two is discussed in the text. The Asian anthropogenic contribution in GEOS-Chem is shown as thin black lines. Figure 7 (bottom) also shows dust concentrations observed at the IMPROVE sites (blue inverted triangles) and simulated by the GEOS-Chem model (thin blue line). Events of transpacific sulfate transport as seen at Midway (see Figure 2) and discussed in the text are highlighted with purple arrows.

Observed dust concentrations are correlated with simulated Asian sulfate contributions ( $R = 0.56$ ), consistent with coincident transport of Asian dust and sulfate across the Pacific. *Van Curen* [2003] found that there was a significant anthropogenic component to the aerosol mass measured at western US IMPROVE sites from 1989 to 1999 on days where Asian dust was observed. *Jaffe et al.* [1999, 2003a] also saw evidence of coincidentally elevated levels of anthropogenic pollutants and aerosols (scattering and absorbing) in Asian plumes transported over North America.

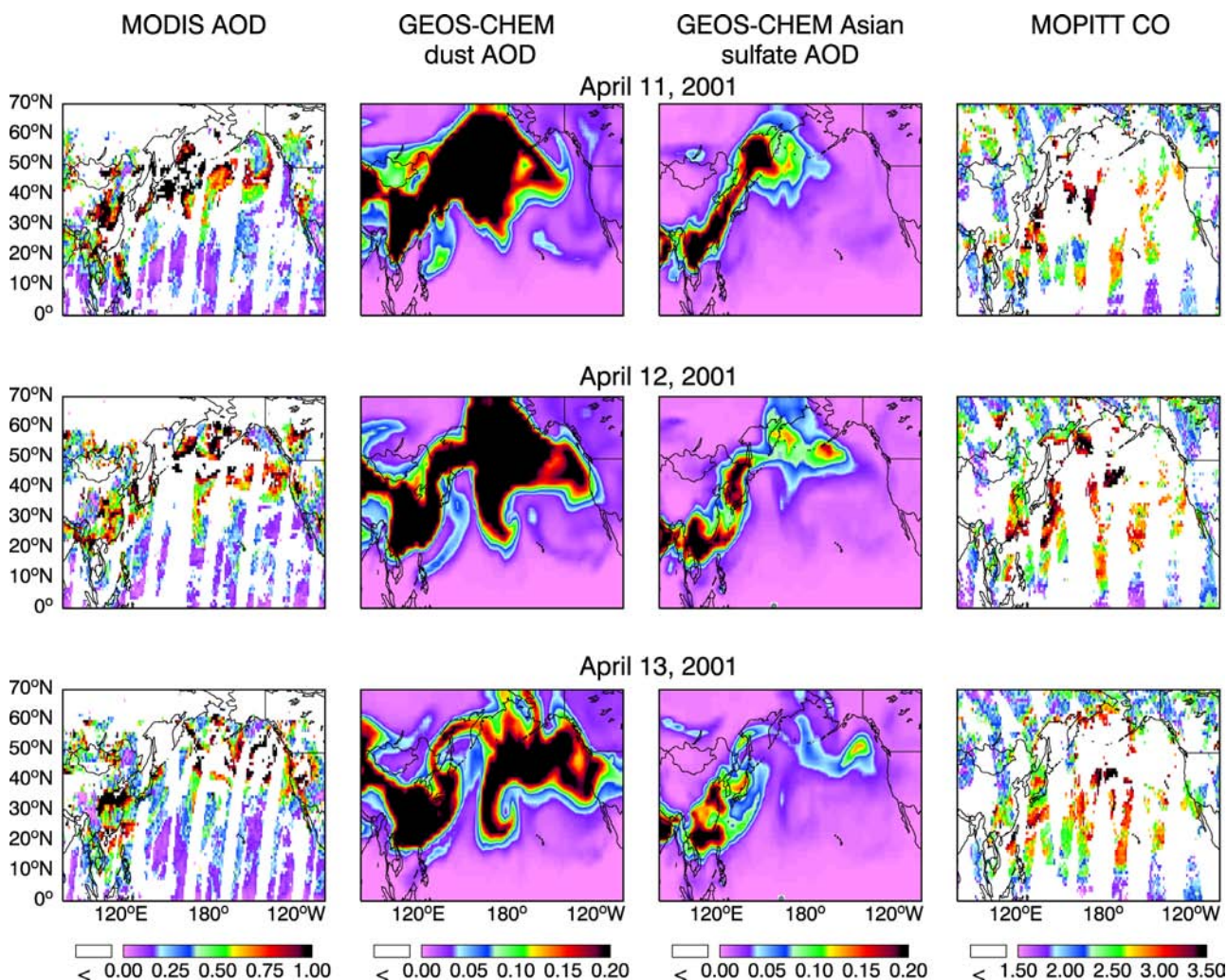
[26] Teasing out the weak signal of transpacific Asian sulfate enhancement in surface air observations is obviously difficult. Except for event 1, the model Asian events in Figure 7 are not associated with clear enhancements in the observations. Nevertheless, we can use statistics of simulated versus observed sulfate for the ensemble of NW IMPROVE sites in Figure 7 (bottom) to place some error bounds on the model estimate of Asian sulfate pollution influence. Asian influence at these sites is low throughout March (Figure 7). Comparison between simulated and observed sulfate every third day for that month (mean bias  $b$  of  $-0.04 \pm 0.18 \mu\text{g m}^{-3}$ , where the uncertainty is the standard deviation of the bias) indicates an unbiased simulation of domestic sulfate. The simulation of total sulfate through the period of maximum Asian influence (10 April to 31 May) is slightly too high ( $b = 0.21 \pm 0.30 \mu\text{g m}^{-3}$ ) with a correlation coefficient  $R = 0.42$ . Zeroing out the Asian contribution from the model simulation of sulfate removes the bias but also greatly weakens the correlation

( $b = 0.01 \pm 0.35 \mu\text{g m}^{-3}$ ,  $R = 0.18$ ). Doubling Asian influence in the model improves the correlation but greatly increases the bias ( $b = 0.41 \pm 0.31 \mu\text{g m}^{-3}$ ,  $R = 0.54$ ). We thus estimate a  $\pm 50\%$  uncertainty on the model results for Asian sulfate enhancements over the NW United States, as larger differences lead to substantial degradations in the quality of the model simulation.

## 6. Linking Surface and Satellite Observations of Transpacific Aerosol Pollution Events

[27] The model time series of Asian sulfate enhancements at the ensemble of IMPROVE sites in the NW United States identifies four prominent Asian events (Figure 7). All were also associated with Asian dust enhancements in the IMPROVE observations, and all were related to events observed by AERONET at Midway a few days before (Figure 2). We examined whether MODIS satellite observations could provide identification and tracking of these events during transport across the Pacific, but found persistent clouds to prevent detection for events 3 and 4. We describe below events 1 and 2.

[28] Event 1 observed in the NW United States on 14–15 April had the highest Asian PM<sub>2.5</sub> dust concentrations for spring 2001 (dust exceeded  $10 \mu\text{g m}^{-3}$  at several IMPROVE sites). Sulfate was also elevated (Figure 7), clearly pointing to the joint transpacific transport of dust and pollution. This was also the largest transpacific transport event seen by MODIS in 2001. Figure 8 shows the MODIS observation of this event progressing across the



**Figure 8.** Transpacific dust event of April 2001 (event 1 in Figures 2 and 7). MODIS AODs (550 nm) are compared to the GEOS-Chem dust and sulfate AODs and to the CO columns observed by MOPITT ( $\times 10^{18}$  molecules  $\text{cm}^{-2}$ ). Color scales are saturated at maximum values.

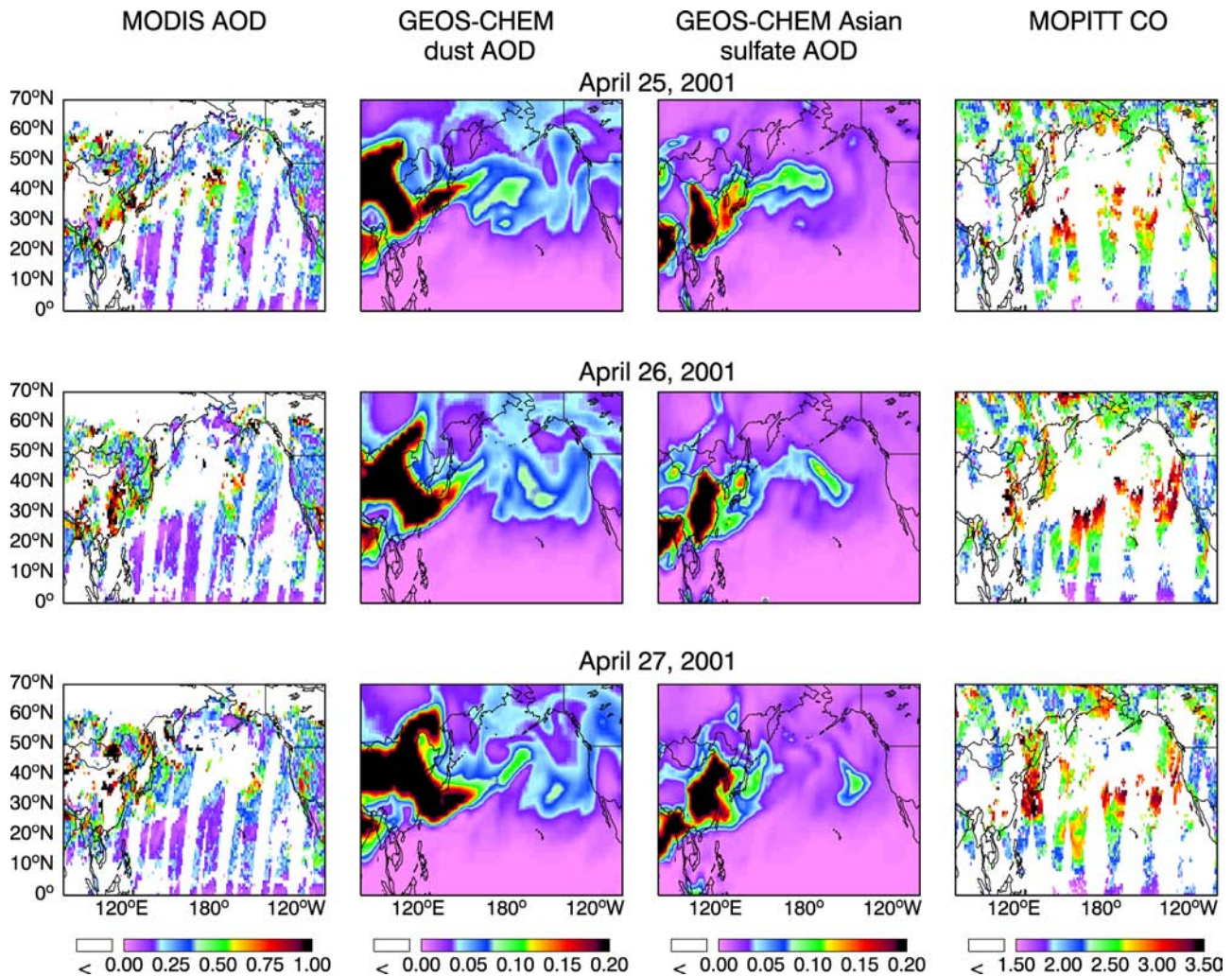
Pacific, along with the simulated contributions of dust and sulfate. As pointed out in section 4, the MODIS AOD bias precludes quantitative comparison, and we focus here on comparing patterns. The dust originated from a major dust storm in the Gobi desert, and the transpacific influence of this dust on sites in both the western and eastern United States has been documented previously [Thulasiraman *et al.*, 2002; Szykman *et al.*, 2003]. A model study by Takemura *et al.* [2002] found that the contribution of anthropogenic aerosols to the enhanced AODs observed by SeaWIFS and at AERONET sites in the northwestern Pacific during this event was comparable to the dust enhancement. The IMPROVE observations and GEOS-Chem simulation also indicate a strong Asian sulfate pollution component transported with the dust. Elevated CO observed by MOPITT accompanying this event (Figure 8) corroborates the strong pollution signature. The pattern of enhanced AODs observed by MODIS matches the combined dust and sulfate AODs simulated in GEOS-Chem, further supporting the model simulation of transpacific transport.

[29] The late April event 2 had a weaker dust signal in both model and observations but comparable Asian sulfate influence according to GEOS-Chem (Figure 7). The temporal evolution of MODIS AODs and MOPITT CO over the Pacific for that event is shown in Figure 9. Despite widespread clouds it is clear that the simulated location of aerosol export, and particularly the large sulfate contribution, corresponds well with the MODIS observations. In this event the contribution of Asian sulfate to the simulated AODs exceeded that of dust according to the model. MOPITT observations indicate high levels of CO over the Pacific coincident with the observed aerosol transport.

**7. Conclusions**

[30] We used a global CTM (GEOS-Chem) simulation of MODIS AOD satellite observations over the North Pacific, and of remote surface air observations in the northwestern United States from the IMPROVE network, to better understand the mechanisms for transpacific transport of Asian aerosol pollution and quantify its impact on North





**Figure 9.** Transpacific sulfate event of April 2001 (event 2 in Figures 2 and 7). MODIS AODs (550 nm) are compared to the GEOS-Chem dust and sulfate AODs and to the CO columns observed by MOPITT ( $\times 10^{18}$  molecules  $\text{cm}^{-2}$ ). Color scales are saturated at maximum values.

American air quality. Our work was motivated by a previous GEOS-Chem study [Park *et al.*, 2004] that found transpacific Asian sulfate to have important implications for current regulations to protect visibility in U.S. national parks. There was a need to test this result as well as to gain knowledge of the processes involved.

[31] The MODIS observations of AODs over the Pacific display the known spring maximum in transpacific aerosol transport and show that its impact on North America is mainly in the 40–55°N latitudinal band. Transport of CO from Asia observed by the MOPITT satellite instrument extends to more southerly latitudes because of export from biomass burning, which appears to be more important for CO than for aerosols. The patterns and timing of export from Asia and transport across the Pacific observed by MODIS are well simulated by the GEOS-Chem CTM, but the model AODs are lower by a factor of two on average. Surface-based observations of AODs from AERONET sites at Midway and Hawaii indicate that this disparity is at least partly due to MODIS retrieval error.

[32] The model indicates that transpacific transport of Asian anthropogenic aerosols is mainly restricted to the

lower free troposphere above the boundary layer (900–700 hPa), unlike anthropogenic CO which is transported throughout the lower-mid troposphere. The relatively low altitude of transpacific aerosol transport facilitates contact with the elevated terrain in the northwestern United States. Simulated springtime Asian enhancements of sulfate in surface air for this region are  $0.16 \mu\text{g m}^{-3}$ , with 24-hour maxima averaging  $0.60 \mu\text{g m}^{-3}$  and values up to  $1.5 \mu\text{g m}^{-3}$  in Washington state.

[33] We analyzed the spring 2001 observations of sulfate at IMPROVE sites in the NW United States for evidence of transpacific Asian pollution and to quantify this influence. The sulfate concentrations observed at these sites average  $1.04 \mu\text{g m}^{-3}$  on the days corresponding to the model 24-hour maximum of local Asian influence, as compared to  $0.69 \mu\text{g m}^{-3}$  for the seasonal mean. A similar analysis for nitrate and OC aerosol shows no significant Asian pollution enhancement. Asian dust events observed at these sites are correlated with Asian sulfate enhancements, indicating the coincident transport of pollution and dust across the Pacific. Four particularly strong events of transpacific aerosol transport are seen in the AERONET record at Midway Island and



subsequently in the model simulation and the observations at the IMPROVE surface sites. Two of these events can be tracked across the Pacific by the MODIS observations, but the other two cannot because of clouds. We find that clouds severely limit the use of satellite observations for detecting transpacific transport of aerosols.

[34] Comparing the simulated and observed time series at the IMPROVE sites in the NW United States suggests a 50% uncertainty in the model estimates of Asian sulfate enhancements in surface air in the United States, i.e., a mean value of  $0.16 \pm 0.08 \mu\text{g m}^{-3}$  for spring 2001 in that region. Extrapolating this relative error estimates to the previously reported results of Park *et al.* [2004] implies a mean annual Asian sulfate enhancement at surface sites in the United States of  $0.10 \pm 0.05 \mu\text{g m}^{-3}$  in the west and  $0.09 \pm 0.4 \mu\text{g m}^{-3}$  in the east. In comparison, the EPA Regional Haze Rule sets a sulfate concentration objective of  $0.09 \mu\text{g m}^{-3}$  in the west and  $0.17 \mu\text{g m}^{-3}$  in the east to guide domestic regulation of emissions aimed at improving visibility in wilderness areas. Our results lend confidence to the findings of Park *et al.* [2004] that transpacific pollution must be considered in setting visibility standards, particularly if sulfur emissions from Asia continue to increase as projected [Streets and Waldhoff, 2000].

[35] We still need better understanding of the possible contribution of OC aerosols to transpacific pollution transport. Observations by Jaffe *et al.* [2005] on the U.S. west coast indicate higher OC than sulfate enhancements in air masses originating from Asia. Our analysis of the IMPROVE data does not identify such an enhancement but this could be due to a relatively large North American natural background [Park *et al.*, 2003]. Aircraft observations off the coast of Japan in spring show a  $1\text{--}4 \mu\text{g m}^{-3}$  OC aerosol background in the free troposphere [Heald *et al.*, 2005] that the model does not capture. Such high levels of OC in the free troposphere would contribute a significant subsiding background to surface air concentrations in the United States, though there is no evidence at present that this background would be anthropogenic.

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