# Description of the NASA GEOS Composition Forecast Modeling System GEOS-CF v1.0

Christoph A. Keller[1], K. Emma Knowland[1], Bryan N Duncan[2], Junhua Liu[1], Daniel C Anderson[1], Sampa Das[2], Robert A Lucchesi[3], Elizabeth W Lundgren[4], Julie M. Nicely[2], Jon Eric Nielsen[5], Lesley E. Ott[2], Emily Saunders[3], Sarah A. Strode[6], Pamela A Wales[7], Daniel J. Jacob[4], and Steven Pawson[2]

[1]Universities Space Research Association
[2]NASA Goddard Space Flight Center
[3]Science Systems and Applications, Inc
[4]Harvard University
[5]SSAI at NASA Goddard Space Flight Center
[6]Universities Space Research Assocaition
[7]University of Maryland, College Park

November 22, 2022

## Abstract

The Goddard Earth Observing System composition forecast (GEOS-CF) system is a high-resolution (0.25 degree) global constituent prediction system from NASA's Global Modeling and Assimilation Office (GMAO). GEOS-CF offers a new tool for atmospheric chemistry research, with the goal to supplement NASA's broad range of space-based and in-situ observations and to support flight campaign planning, support of satellite observations, and air quality research. GEOS-CF expands on the GEOS weather and aerosol modeling system by introducing the GEOS-Chem chemistry module to provide analyses and 5-day forecasts of atmospheric constituents including ozone ($O_3$), carbon monoxide (CO), nitrogen dioxide ($NO_2$), and fine particulate matter (PM2.5). The chemistry module integrated in GEOS-CF is identical to the offline GEOS-Chem model and readily benefits from the innovations provided by the GEOS-Chem community.

Evaluation of GEOS-CF against satellite, ozonesonde and surface observations show realistic simulated concentrations of $O_3$, $NO_2$, and CO, with normalized mean biases of -0.1 to -0.3, normalized root mean square errors (NRMSE) between 0.1-0.4, and correlations between 0.3-0.8. Comparisons against surface observations highlight the successful representation of air pollutants under a variety of meteorological conditions, yet also highlight current limitations, such as an overprediction of summertime ozone over the Southeast United States. GEOS-CF v1.0 generally overestimates aerosols by 20-50% due to known issues in GEOS-Chem v12.0.1 that have been addressed in later versions.

The 5-day hourly forecasts have skill scores comparable to the analysis. Model skills can be improved significantly by applying a bias-correction to the surface model output using a machine-learning approach.

1
2
# **Description of the NASA GEOS Composition Forecast Modeling System GEOS-CF v1.0**

3 **Christoph A. Keller[1], K. Emma Knowland[1,2], Bryan N. Duncan[1], Junhua Liu[1,2], Daniel C.**
4 **Anderson[1,2], Sampa Das[1,2], Robert A. Lucchesi[1,3], Elizabeth W. Lundgren[4], Julie M.**
5 **Nicely[1,5], Eric Nielsen[1,3], Lesley E. Ott[1], Emily Saunders[1,3], Sarah A. Strode[1,2], Pamela A.**
6 **Wales[1,2], Daniel J. Jacob[4], Steven Pawson[1]**

7 [1]NASA Goddard Space Flight Center, Greenbelt, MD, USA

8 [2]Universities Space Research Association, Columbia, MD, USA

9 [3]Science Systems and Applications, Inc., Lanham, MD, USA

10 [4]School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA

11 [5]Science Systems and Applications, Inc., Lanham, MD, USA

12

13 Corresponding author: Christoph Keller ([christoph.a.keller@nasa.gov](mailto:christoph.a.keller@nasa.gov))

14 **Key Points:**

15
16 - GEOS-CF is a new modeling system that produces global forecasts of atmospheric composition at $25km^2$ horizontal resolution.

17
18 - GEOS-CF model output is freely available and offers a new tool for academic researchers, air quality managers, and the public.
19

**Abstract**

The Goddard Earth Observing System composition forecast (GEOS-CF) system is a high-resolution (0.25 degree) global constituent prediction system from NASA's Global Modeling and Assimilation Office (GMAO). GEOS-CF offers a new tool for atmospheric chemistry research, with the goal to supplement NASA's broad range of space-based and in-situ observations and to support flight campaign planning, support of satellite observations, and air quality research. GEOS-CF expands on the GEOS weather and aerosol modeling system by introducing the GEOS-Chem chemistry module to provide analyses and 5-day forecasts of atmospheric constituents including ozone ($O_3$), carbon monoxide (CO), nitrogen dioxide ($NO_2$), and fine particulate matter (PM2.5). The chemistry module integrated in GEOS-CF is identical to the offline GEOS-Chem model and readily benefits from the innovations provided by the GEOS-Chem community.

Evaluation of GEOS-CF against satellite, ozonesonde and surface observations show realistic simulated concentrations of $O_3$, $NO_2$, and CO, with normalized mean biases of -0.1 to -0.3, normalized root mean square errors (NRMSE) between 0.1-0.4, and correlations between 0.3-0.8. Comparisons against surface observations highlight the successful representation of air pollutants under a variety of meteorological conditions, yet also highlight current limitations, such as an overprediction of summertime ozone over the Southeast United States. GEOS-CF v1.0 generally overestimates aerosols by 20-50% due to known issues in GEOS-Chem v12.0.1 that have been addressed in later versions.

The 5-day hourly forecasts have skill scores comparable to the analysis. Model skills can be improved significantly by applying a bias-correction to the surface model output using a machine-learning approach.

**Plain Language Summary**

Accurate forecasting of the compostion of the atmosphere is important for a variety of applications, including air pollution mitigation, support of satellite and other remote-sensing observations, and research applications. Producing such forecasts is computationally expensive due to the complexity of atmospheric chemistry, which interacts with weather on all scales. Here we present the NASA Goddard Earth Observing System composition forecast (GEOS-CF) system, which produces global forecasts of major atmospheric constituents such as ozone ($O_3$), nitrogen dioxide ($NO_2$), and fine particulate matter (PM2.5). On a daily basis, the model tracks the atmospheric concentrations of more than 250 chemical species in more than 55 million model grid cells, computing the interactions between those species using the state-of-the-science GEOS-Chem chemistry model.

We present and in-depth evaluation of the GEOS-CF model through comparison against independent observations. We show how the model captures many observed features of atmospheric composition, such as spatio-temporal variations in air pollution due to changes in pollutant emissions, weather, and chemistry. We also highlight some of the model deficiencies, e.g., with respect to the simulation of aerosol particles. Finally, we demonstrate how surface observations and model data can be combined using machine learning to provide improved local air quality forecasts.

**1 Introduction**

Near real-time information of global atmospheric composition is invaluable for a wide range of applications, including academic research, airborne and satellite mission support, air

64  quality forecasting, disaster management, and ecosystem monitoring. However, the numerical
65  simulation of atmospheric chemistry is computationally expensive because it involves hundreds
66  of species that interact with each other on time scales from milliseconds to years, and the species
67  are also influenced by dynamics across a wide range of spatiotemporal scales. This precludes the
68  inclusion of detailed aerosol and reactive trace gases in standard operational numerical weather
69  prediction (NWP) systems. Instead, real-time simulation of atmospheric composition is typically
70  done within a simplified system in order to reduce the computational burden, e.g., by running the
71  model at reduced horizontal resolution or over a regional domain only, using a simplified
72  representation of atmospheric composition, or by coupling a weather model with an offline
73  chemical transport model (CTM) (e.g., Bhattacharjee et al., 2018; Emmons et al., 2020;
74  Flemming et al., 2015; Marécal et al., 2015).

76       Here we present the NASA Goddard Earth Observing System (GEOS) composition
77  forecast modeling system, GEOS-CF v1.0, which provides global analyses and forecasts of
78  atmospheric composition such as ozone ($O_3$), carbon monoxide (CO), nitrogen dioxide ($NO_2$),
79  and fine particulate matter ($PM_{2.5}$) in near real-time at a horizontal resolution of approximately
80  $25x25$ $km^2$. To our knowledge, GEOS-CF is one of only a few global forecasting systems of
81  atmospheric composition conducted in near real-time. The European Centre for Medium-Range
82  Weather Forecasts (ECMWF) offers 5-day global forecasts of aerosols and trace gases at
83  approximately $40x40$ $km^2$ horizontal resolution through the Copernicus Atmosphere Monitoring
84  Service (CAMS, https://atmosphere.copernicus.eu/global-forecast-plots). The US National
85  Center for Atmospheric Research (NCAR) conducts 10-day global forecasts at approximately
86  $100x100$ $km^2$ horizontal resolution based on offline simulations of the Model for Ozone and
87  Related chemical Tracers (MOZART) Chemistry Mechanism in the Community Earth System
88  Model Version 2 (CESM2) (Emmons et al., 2020) driven by GEOS meteorological forecasts
89  (https://www2.acom.ucar.edu/acresp/forecasts-and-near-real-time-nrt-products). Finally, the
90  Finnish Meteorological Institute provides daily 4-day global forecasts of atmospheric
91  composition at approximately $35x35$ $km^2$ resolution using the System for Integrated modeLling
92  of Atmospheric coMposition (SILAM v5.7, http://silam.fmi.fi/). The atmospheric composition
93  forecasts from these models can vary considerably due to differences in the underlying
94  meteorological fields, observational constraints, chemical mechanisms, or assumptions about
95  pollutant emissions. The uncertainties associated with these processes can be difficult to quantify
96  from a single model simulation alone, and the availability of multiple, independently developed
97  models offers great potential to provide better air quality information through combination of a
98  suite of models (Marécal et al., 2015). GEOS-CF offers such an independent composition
99  forecast, thus complementing the existing suite of global composition forecasting systems by
100  providing global 5-day forecasts of atmospheric composition using the GEOS-Chem atmospheric
101  chemistry module (http://www.geos-chem.org) within GEOS from January 2018 onward.

103       One of the key aspects of the GEOS-CF system is the full integration of the GEOS-Chem
104  model in the GEOS system, which allows for the simulation of reactive gases and aerosols at the
105  same temporal and spatial resolution as the meteorology (Long et al., 2015; Hu et al., 2018).
106  GEOS-Chem is actively evaluated and developed by a large international research community,
107  and the GEOS-Chem module incorporated in GEOS-CF uses the exact same codebase as the
108  offline CTM. This enables the seamless integration of scientific updates provided by the GEOS-
109  Chem CTM community into GEOS-CF without the need to make any modifications to the source

110  code. The scientific validity of the GEOS-Chem chemistry module within GEOS has been
111  demonstrated by Hu et al. (2018), who show that a one-year global simulation of atmospheric
112  composition at 12.5 km$^2$ produces results consistent with the offline GEOS-Chem model. GEOS-
113  CF is a natural extension of the online GEOS-Chem module embedded in GEOS, with a focus on
114  daily operation and forecast capabilities.
115
116      GEOS-CF is the latest in a series of research and applications products generated by the
117  NASA Global Modeling and Assimilation Office (GMAO), including the GEOS forward
118  processing weather and aerosol system (GEOS FP), GEOS FP for instrument teams (GEOS FP-
119  IT, Lucchesi, 2015), the second Modern-Era Retrospective analysis for Research and
120  Applications (MERRA-2, Gelaro et al., 2017), and the Seasonal to Subseasonal Forecasting
121  System (GEOS-S2S, Borovikov et al., 2019; Molod et al., 2020). It leverages GMAO's model
122  infrastructure and directly builds on a number of development activities centered around the
123  GEOS model, with the goal to extend these forecasting capabilities toward (short-lived) trace
124  gases and aerosols.
125  GEOS is a General Circulation Model (GCM) and Data Assimilation System (DAS) consisting
126  of a suite of model components that can be connected in a modular manner through the Earth
127  System Modeling Framework (ESMF, Hill et al., 2004) and Modeling Analysis and Prediction
128  Layer (MAPL, Suarez et al., 2007) software interface. The model can be configured to run with
129  fully interactive chemistry so that the chemical constituents feed back to the dynamics ('online'),
130  or as an offline model where external meteorological fields are used as input. A hybrid approach
131  is the 'replay' feature, where the model dynamics are nudged toward pre-computed analysis
132  fields (from a previous DAS simulation) in a way that is consistent with the internal physics of
133  the model (Orbe et al., 2017). This approach is particularly useful for chemistry simulations as it
134  sidesteps the need to conduct a computationally costly meteorological data assimilation cycle.
135  Several chemistry and aerosol modules of varying complexity are available in GEOS (Nielsen et
136  al., 2017), enabling a wide range of applications including: near real-time simulation of aerosols
137  using the GOCART module (Colarco et al. 2010; Randles et al., 2017; Buchard et al., 2017),
138  computationally efficient analysis of stratospheric ozone using parameterized chemistry in
139  combination with 3D-Variational assimilation of satellite observations (Wargan et al., 2015);
140  multi-decade simulation of tropospheric and stratospheric chemistry using the Global Modeling
141  Initiative (GMI) chemistry module (Douglass et al., 2004; Duncan et al., 2007, Strahan et al.,
142  2007); and fully coupled simulation of gas-phase and aerosol chemistry using the GEOS-Chem
143  chemistry module embedded in GEOS (Long et al., 2015).
144
145      In this paper we describe the configuration of GEOS-CF version 1.0 (Section 2) and
146  demonstrate the validity of the produced analyses through comparison against independent
147  observations (Sections 3-4). Model forecasts are evaluated in Section 5 and we discuss how
148  model skill scores can be improved by applying a bias-correction to the surface observations
149  using a machine-learning approach. As described in detail below, the current version of GEOS-
150  CF constitutes a hybrid model between an online weather and chemistry assimilation system and
151  an offline CTM application, with a development pathway toward a fully coupled forecasting
152  system with integrated trace gases and aerosols. Many of its design features were guided by
153  practical considerations as well as computational limitations related to the simulation of
154  atmospheric chemistry.

## 2 Model Description

2.1. General Description

A schematic of the GEOS-CF v1.0 modeling system is provided in Figure 1. GEOS-CF is operated in near real-time, producing a 5-day forecast once a day. The forecast initial conditions are provided by a one-day replay simulation (i.e., 'analysis') constrained by pre-computed meteorological analysis fields. In the v1.0 configuration, the analysis forcings that GEOS-CF uses are GEOS FP-IT meteorological variables and GOCART aerosols providing meteorological feedbacks, and GEOS-FP stratospheric ozone (described below). All other trace gases are integrated without observational constraints. The resulting model conditions at the end of the simulated day also serve as input for the next day's replay step (i.e., restart files). This preserves the model chemical and meteorological state from one forecast cycle to the next, leading to a continuous atmospheric composition archive since January 2018.

GEOS FP-IT is a 'frozen' model system that is comparable to MERRA-2 but - unlike MERRA-2 - is available in near real-time to support retrievals by satellite instrument teams. The GEOS model in general, and version of the GEOS GCM used by GEOS FP-IT (v5.12.4) in particular, has shown to be well suited for atmospheric chemistry applications as it realistically captures features critical to atmospheric composition, such as the seasonal climate of moisture and temperature and large-scale transport of constituents (e.g., Pawson et al., 2007; Douglass et al., 2014; Oman and Douglass, 2014; Molod et al., 2015; Gelaro et al., 2017).

GEOS-CF v1.0 uses a model physics package that is similar to MERRA-2 and GEOS FP-IT, consisting of the GEOS atmospheric model, version 5, described in Rienecker et al. (2008) with updates as described in Molod et al. (2012) and Molod et al. (2015). The model uses the finite-volume dynamical core of Lin (2004) with a cubed sphere grid discretization to avoid grid-cell singularities (Putman and Lin, 2007). It is run at cubed-sphere c360 horizontal resolution (roughly equivalent to 0.25° x 0.25°) and 72 hybrid-eta levels from the surface to 0.01 hPa. Model physics includes parameterizations for moist processes, radiation, turbulent mixing, land-surface processes, and gravity wave drag. The moist module contains parameterization of convection using the Relaxed Arakawa-Schubert scheme (Moorthi & Suarez, 1992), and the single-moment parameterization for large-scale precipitation and cloud cover described in Bacmeister et al. (2006). The radiation module includes parameterization for long-wave (Chou 1990, 1992) and short-wave radiation processes (Chou and Suarez 1994). Turbulence is parameterized using the gradient Richardson number in the free atmosphere and the Lock scheme (Lock et al., 2000) interfaced with the scheme of Louis and Geleyn (1982) in the boundary layer. Exchange of heat, moisture and momentum between land, atmosphere, and ocean or sea ice surfaces are parameterized using Monin-Obukhov similarity theory (Helfand and Schubert, 1995, Molod et al., 2013), and the gravity wave drag parameterization contains orographic (McFarlane, 1987) and non-orographic (Garcia & Boville, 1994) waves.

In the GEOS-CF system, the GEOS physics components are coupled to the GOCART aerosol component to provide consistent physics with the GEOS FP-IT meteorology. The GEOS-Chem chemistry module is run "passively" to provide coupled aerosol-oxidant chemistry in the troposphere and stratosphere (Hu et al., 2018). The GOCART aerosols, constrained during the replay step by aerosol optical depth (AOD) observations from the Moderate Resolution Imaging Spectroradiometer (MODIS) aboard the Terra and Aqua satellites (Randles et al., 2017), are used

to compute the feedback between aerosols and dynamics. This ensures consistency between GEOS-CF and the GEOS FP-IT analysis fields. Currently, there is no chemical coupling between GOCART and GEOS-Chem, and no observations are directly assimilated into GEOS-CF. However, stratospheric ozone in GEOS-Chem is nudged towards ozone produced by GEOS FP, which is constrained by ozone measurements from the Microwave Limb Sounder (MLS), Ozone Monitoring Instrument (OMI), and NASA's Ozone Mapping and Profiler Suite (OMPS) and produces a realistic analysis of ozone in the stratosphere (Wargan et al., 2015; 2020). In addition, near real-time MODIS observations of fire radiative power are used to constrain fire emissions, as produced by the Quick Fire Emissions Dataset (QFED) (Darmenov and Da Silva, 2015).

All computations are conducted on the Discover supercomputing cluster of the NASA Center for Climate Simulation (NCCS). Run on 3510 Intel Xeon Haswell processor cores, the one-day analysis and 5-day forecast takes approximately 8.5 wall-clock hours. GEOS-CF analysis and forecast output includes chemistry and meteorology "surface" output every 15 minutes as well as hourly-average and instantaneous fields for surface, column-average, and 3-dimensional model output. The model output is publicly available at https://gmao.gsfc.nasa.gov/weather_prediction/GEOS-CF/data_access/ in the form of on-demand figures or through access to the model output (in netCDF data format) via Hypertext Transfer Protocol (HTTP) file download or through the Open-source Project for a Network Data Access Protocol (OPeNDAP) remote access tool. The full 5-day model forecast output is publicly available for a duration of 14 days. Given the growing interest in air quality forecasting applications, the model forecasts for a selection of surface air pollutants are made available on the public portal indefinitely. Full details on available output and data access are available in the GEOS-CF File Specification document (Knowland et al., 2020).
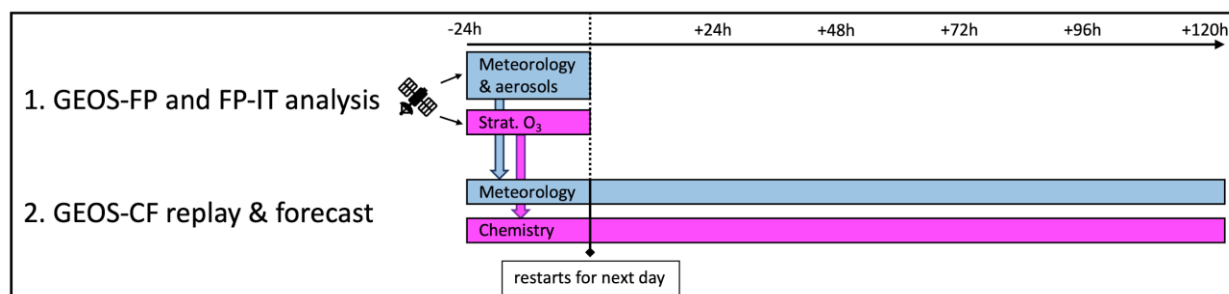


**Figure 1.** Schematic of the GEOS-CF modeling system approach, consisting of one day analysis and 5-day forecast. This combination of simulations is conducted on a daily basis.

2.2. Chemistry

GEOS-CF v1.0 uses the continually updated standard version of the GEOS-Chem chemistry module to simulate coupled aerosol-oxidant chemistry in the troposphere and stratosphere. The current version 12.9.2 of GEOS-Chem has been implemented into GEOS-CF as of this writing, but results are presented here for version 12.0.1 in order to have a two-year record for comparison to observations. GEOS-Chem is ESMF-compliant and its chemistry module is implemented here as an ESMF gridded component of GEOS, as described in Long et al. (2015) and Hu et al. (2018).

The gas-phase chemistry scheme includes detailed tropospheric chemistry of $HO_x$, $NO_x$, $BrO_x$, volatile organic compounds (VOC), and $O_3$, as originally described by Bey et al. (2001), with addition of halogen chemistry by Parrella et al. (2012) and Sherwen et al. (2016) plus updates to

238  isoprene oxidation as described by Mao et al. (2013) and Miller et al. (2017). Stratospheric
239  chemistry is fully coupled with tropospheric chemistry through the Unified tropospheric-
240  stratospheric Chemistry eXtension (UCX, Eastham et al. (2014)) and extends to the top of the
241  atmosphere. Photolysis rates are computed by GEOS-Chem using the Fast-JX code (Bian and
242  Prather, 2002). The gas-phase mechanism comprises 250 chemical species and 725 reactions and
243  is solved using the Kinetic PreProcessor KPP Rosenbrock solver (Sandu and Sander, 2006). The
244  aerosol simulation includes sulfate-nitrate-ammonia chemistry (Park et al., 2004), black carbon
245  (Park et al., 2003; Wang et al., 2014), organic aerosols (Marais et al., 2016), mineral dust (Fairlie
246  et al., 2007; Ridley et al., 2012), and sea salt aerosols (Jaeglé et al., 2011). Aerosol and gas-phase
247  chemistry interact through gas-aerosol partitioning (Fountoukis and Nenes, 2007; Pye et al.,
248  2009), heterogeneous chemistry on aerosol surface (Evans and Jacob, 2005; Mao et al., 2013),
249  and aerosol impacts on photolysis (Martin et al., 2003). Methane concentrations are prescribed as
250  monthly mean surface concentrations, spatially interpolated from NOAA GLOBALVIEW flask
251  data (Dlugokencky et al., 1995).

252      2.3. Emissions and Deposition

253      The dry deposition scheme in GEOS-Chem is based on the resistance-in-series model of
254  Wesely (1989). Wet deposition of aerosols and soluble gases includes scavenging in convective
255  updrafts, in-cloud rainout, and below-cloud washout (Liu et al., 2001; Amos et al., 2012). All
256  emission calculations are done using the Harmonic Emissions Component HEMCO v2.1.009
257  (Keller et al., 2014). Table 1 summarizes the emission configuration used by GEOS-CF v1.0.
258  Anthropogenic emissions are broken down into hourly values using sector-specific day-of-week
259  and diurnal scale factors (van der Gon et al., 2011). In addition, an annual gridded scale factor
260  based on the Open-source Data Inventory for Anthropogenic $CO_2$ (ODIAC; Oda et al., 2017) is
261  applied to the anthropogenic emissions of CO.

262

263  **Table 1.** *Emissions used by GEOS-CF.*

| Description | Reference | Comments |
|---|---|---|
| *Offline inventories* | | |
| Anthropogenic NO, CO, black carbon (BC), organic carbon (OC), Ammonia ($NH_3$) | HTAP v2.2 (Janssens-Maenhout et al., 2015) | Global except Africa |
| Anthropogenic $SO_2$ | OMI-HTAP (Liu et al., 2018) | Global except Africa |
| Anthropogenic VOCs | RETRO (Schultz et al., 2008) | Global except Africa |
| Anthropogenic NO, CO, $SO_2$, BC, OC, $NH_3$, VOCs | DICE-Africa (Marais and Wiedinmyer, 2016) | Africa |
| Arctic seabird $NH_3$ | Croft et al. (2016) | |
| Volcanic $SO_2$ | Carn (2019) | 5% of the sulfur emitted as $SO_4$ |
| Aircraft $NO_x$, CO, $SO_2$, VOCs, BC, OC | AEIC (Stettler et al., 2011) | |
| *Dynamic emissions* | | |
| Biogenic VOCs | MEGAN v2.1 (Guenther et al., 2012) | |
| Biomass burning $NO_x$, CO, $SO_2$, VOCs, BC, OC | QFED v2.5 (Darmenov and da Silva, 2015) | 35% emitted between 3.5 and 5.5 km altitude (Fischer et al., 2014). |
| Lightning $NO_x$ | Murray et al., 2012 | |
| Soil $NO_x$ | Hudman et al., 2012 | |
| Soil dust | Zender et al., 2003 | |

| Sea salt aerosols | Gong, 2003; Jaeglé et al., 2011 | |
| Oceanic DMS, $CH_2O$, $C_3H_6O$ | Johnson, 2010; Nightingale et al., 2000 | |
| Oceanic iodine | Carpenter et al., 2013 | |

## 3 Observations used for Model Evaluation

GEOS-CF is intended to supplement NASA's broad range of space-based and in-situ observations, providing a new tool for researchers, government scientists, and air quality managers. We therefore focus our evaluation on the species most pertinent to these applications, including $O_3$, $NO_2$, CO, and aerosols. These species are also constantly evaluated by the broader GEOS-Chem community using the standard CTM simulations (e.g., Hu et al., 2017, 2018; Travis et al., 2019). However, it should be noted that GEOS-CF simulations can differ from standard GEOS-Chem simulations due to the higher horizontal resolution and differences in the emission inputs, in particular wildfire emissions (Hu et al., 2018).

We first evaluate the ability of the GEOS-CF analysis to provide a realistic representation of atmospheric composition based on the hourly-average analysis fields (Section 4). Differences between the 5-day model forecasts and the model analysis are discussed in Section 5.

The data sets used for model validation are summarized in Table 2. Briefly, we evaluate the global tropospheric distribution of $O_3$ against ozonesonde observations obtained from the World Ozone and Ultraviolet Data Center (WOUDC, http://www.woudc.org), $NO_2$ against the OMI NASA standard tropospheric column $NO_2$ product v4.0 (Lamsal et al., 2020), CO total columns against satellite retrievals from the Measurements Of Pollution In The Troposphere (MOPITT) V8 (Deeter et al., 2019), and AOD against the Aerosol Robotic Network (AERONET) level 2.0 daily average data from the version 3 data product (https://aeronet.gsfc.nasa.gov/; Giles et al., 2019). In addition, we compare simulated surface concentrations against in-situ observations from the Global Atmospheric Watch (GAW) World Data Center for Greenhouse Gases (WDCGG, https://gaw.kishou.go.jp/) and World Data Centre for Reactive Gases (WDCRG, https://www.gaw-wdcrg.org/), as well as observations from the OpenAQ database (https://openaq.org). For $NO_2$, we omitted mountainous GAW sites given that the model resolution of 25x25 $km^2$ is not high enough to resolve the fine-scale vertical gradients around mountain slopes. On average, the depth of the GEOS-CF model surface layer is 130 meters and we use this value without attempting to adjust for sub-grid vertical gradients (Travis et al., 2019). All aerosol evaluation is based on the GEOS-Chem aerosols, and model $PM_{2.5}$ is calculated for a relative humidity (RH) of 35%. The validation covers the time period from the start of the GEOS-CF data record on Jan 1, 2018 to Dec 31, 2019.

Figure 2 shows the global distribution of all in-situ observations used in the evaluation. The GAW observation sites are located away from the major pollution sources, representing the global background concentrations. In contrast, most observations available on the OpenAQ platform are located in densely populated areas, providing information about local air pollution. For our analysis, we only include OpenAQ locations with at least 80% of data coverage during the 2-year period. The OpenAQ and AERONET observations are further grouped into six geographical regions, as shown in Figure 2. This subset of observations provides good coverage for North America, Europe, and Eastern Asia (especially China) but a limited view of other

304 regions such as South America or Africa. For those under-represented regions, our analysis relies
305 heavily on the global daily satellite observations.

306

307 **Table 2.** *Overview of observation data sets used for GEOS-CF model validation*

| Description | Species | # sites | Reference |
| --- | --- | --- | --- |
| Ozonesonde | $O_3$ | 24 | Thompson et al., 2017 |
| NASA OMI $NO_2$ v4.0 | Tropospheric $NO_2$ | global | Lamsal et al., 2020 |
| MOPITT v8 | Total CO | global | Deeter et al., 2019 |
| AERONET | AOD at 550nm | 195 | Giles et al., 2019 |
| GAW WDCGG | CO | 54 | https://gaw.kishou.go.jp/ |
| GAW WDCRG | $O_3$, $NO_2$ | 48 ($O_3$), 6 ($NO_2$) | https://www.gaw-wdcrg.org/ |

308



309
310 **Figure 2.** Overview of observation sites used for model validation. Black boxes show the six
311 regions used for aggregation of OpenAQ and Aeronet observations.

## 4 Evaluation of Model Analysis

### 4.1. Model Skill Scores

314     We first highlight the model skill scores against all surface observations before
315 discussing the individual species in more detail below. Figure 3 shows monthly model skill
316 scores for $O_3$, $NO_2$, CO, and $PM_{2.5}$ and/or AOD, aggregated by observation type. The skill scores

317 used are the normalized mean bias (NMB), normalized root mean square error (NRMSE), and
318 Pearson correlation coefficient (R):

319

320 $$NMB = \frac{\Sigma_{n=1}^{N}(y_n - o_n)}{\bar{o}} \qquad (1)$$

321 $$NRMSE = \frac{\sqrt{\frac{1}{N}\Sigma_{n=1}^{N}(y_n - o_n)^2}}{o_{0.95} - o_{0.05}} \qquad (2)$$

322

323 $$R = \frac{\Sigma_{n=1}^{N}(y_n - \bar{y})(o_n - \bar{o})}{\sqrt{\Sigma_{n=1}^{N}(y_n - \bar{y})^2}\sqrt{\Sigma_{n=1}^{N}(o_n - \bar{o})^2}} \qquad (3)$$

324

325 where $y_n$ denotes an indidivual model estimate, $\bar{y}$ is the model average, $o_n$ is the observation
326 associated with $y_n$, and $\bar{o}$ is the observation average; $o_{0.05}$ and $o_{0.95}$ are the 5th and 95th percentile,
327 respectively, of the observations sample, and N is the total number of hourly observation-model
328 pairs.
329 　　For hourly average surface $O_3$, the model shows a normalized mean annual bias of -0.015
330 compared to the GAW sites, with little seasonal variability (Figure 3). The GAW sites are
331 typically located in remote locations and the small model bias indicates that the model
332 adequately captures atmospheric background $O_3$ throughout the year. This is confirmed by the
333 low NRMSE of 0.12 and the correlation coefficient of 0.66, again with small seasonal variability.
334 Compared to the GAW sites, the simulated $O_3$ shows weaker skill scores relative to the OpenAQ
335 observations with an NMB of 0.13, an NRMSE of 0.43 and R of 0.49. As further discussed
336 below, the OpenAQ comparisons highlight a pronounced model overestimation of surface $O_3$
337 during the summer months, especially over North America.
338 For $NO_2$ (second column of Figure 3), the normalized model bias is -0.06 compared against the
339 GAW sites, and -0.30 against the OpenAQ sites. The model shows a strong seasonal change in
340 the NMB at the GAW sites, where the model on average overpredicts wintertime $NO_2$ by up to
341 30% but underpredicts spring and early summer concentrations by a similar amount. The
342 OpenAQ bias shows the opposite direction with a reduction of the negative bias from -0.4 during
343 winter to -0.25 during summer. The NRMSE is 0.15 at the GAW sites and 0.48 at the OpenAQ
344 sites, similar to $O_3$. The R values for $NO_2$ are the lowest for all analyzed species, with an average
345 score of 0.27 and 0.29 at the GAW and OpenAQ sites, respectively. Apart from potential $NO_2$
346 observation interference with other nitrogen compounds (Winer et al., 1974; Grosjean and
347 Harrison, 1985; Steinbacher et al., 2007), we attribute this to the short atmospheric lifetime of
348 $NO_2$, which makes it challenging for the model to capture the hourly variability in surface $NO_2$
349 in full detail.
350 The simulated CO compares well against the 54 GAW sites, with an NMB of -0.036, an NRMSE
351 of 0.088 and R of 0.74. The model error is largest during the NH summer months, with a
352 decrease in NMB to -0.14, an increase in NRMSE to 0.18, and a reduction of R to 0.5.
353 The model comparisons against AERONET AOD and OpenAQ $PM_{2.5}$ observations reveal a
354 systematic model overestimation of aerosol concentrations in GEOS-CF, with an average NMB
355 of 0.61 for AOD and 1.6 for surface $PM_{2.5}$. Known model issues in the treatment of model
356 emissions and wet scavenging contribute to these biases, as will be discussed in more detail
357 below. In addition, some $PM_{2.5}$ observations on the OpenAQ platform represent dry particulate
358 matter while model $PM_{2.5}$ assumes 35% RH, which also contributes to the mismatch. At the
359 AERONET sites, the model shows a good correlation score of 0.71 and the NMRSE of 0.15 is
360 comparable to that of other species at background sites, indicating that the model captures the

361 relative changes in AOD well despite the high model bias. For surface $PM_{2.5}$, the average model
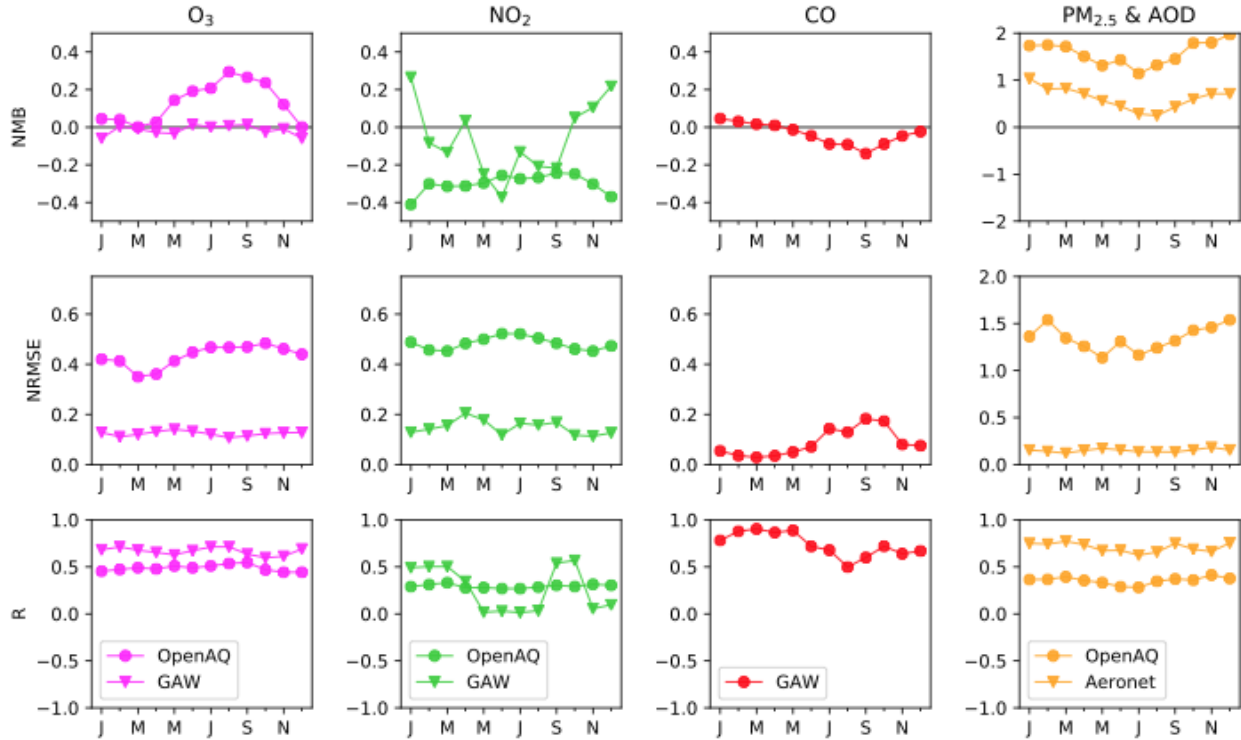362 NRMSE is 1.34 and R skill score is 0.35.
363



364
365 **Figure 3.** Overview of NMB, NRMSE and R for $O_3$, $NO_2$, CO, and $PM_{2.5}$ (OpenAQ only) and
366 AOD (Aeronet only). Shown are the monthly skill score means aggregated by observation type.
367 Note the different scale for $PM_{2.5}$ and AOD.

368        4.2. Ozone

369        Comparisons of GEOS-CF against ozonesondes and GAW surface observations show
370 that GEOS-CF reproduces well-known features of background surface ozone, such as the local
371 maxima in the Northern Hemisphere (NH) during spring.
372        Figure 4 shows annual mean (2018-2019) vertical ozone profiles as observed by
373 ozonesondes at 24 locations across the world (see Figure 2 for locations), compared against
374 corresponding GEOS-CF model concentrations. While GEOS-CF generally captures the
375 observed vertical structure of ozone, the model tends to underestimate free tropospheric ozone
376 (approx. 800-300 hPa) over the NH midlatitudes. For these profiles (e.g., De Bilt, Payerne,
377 Madrid, Tateno), the simulated vertical gradient is much less pronounced than observed and the
378 model consistently underestimates ozone concentrations. This pattern is consistent with previous
379 comparisons of the online GEOS-Chem module against observations (Hu et al., 2018) and
380 implies two potential model errors: (1) excessive vertical mixing resulting in an overestimation
381 in the lower altitudes and underestimation at higher altitudes, as evident over Payerne,
382 Yarmouth, and Madrid (Figure 4); and (2) an underestimation of lightning $NO_x$ over the NH
383 midlatitudes, resulting in an underestimation of ozone production in the upper troposphere. The
384 global lightning $NO_x$ source in GEOS-CF is 5.9 Tg N $yr^{-1}$, in good agreement with other studies
385 (Schumann and Huntrieser, 2007) and the 6.0 Tg N $yr^{-1}$ reported for the GEOS-Chem CTM
386 (Murray et al., 2012). However, due to the real-time nature of the system, the GEOS-CF

387    lightning parameterization uses the unconstrained cloud top height parameterization (Price and
388    Rind, 1992; 1993; 1994) and does not apply time-dependent, regional redistribution factors based
389    on Lightning Imaging Sensor (LIS) and the Optical Transient Detector (OTD) satellite
390    observations as is standard in GEOS-Chem. As described in Murray et al. (2012), this results in
391    an underestimation of simulated lightning flash rates over the Northern extratropics, which is
392    likely one of the main reasons for the model underestimation of ozone in the NH upper
393    troposphere. We also note that the model vertical resolution is approx. 500m (20 hPa) in the mid-
394    to upper troposphere, which might be insufficient to resolve the strong vertical gradients across
395    the tropopause boundary and contribute to the model-observation mismatches in the upper
396    troposphere. Over the Southern Hemisphere (SH), the simulated ozone profiles are in good
397    agreement with the ozonesonde observations and show an improved $O_3$ distribution compared to
398    the offline GEOS-Chem model (Hu et al., 2018).
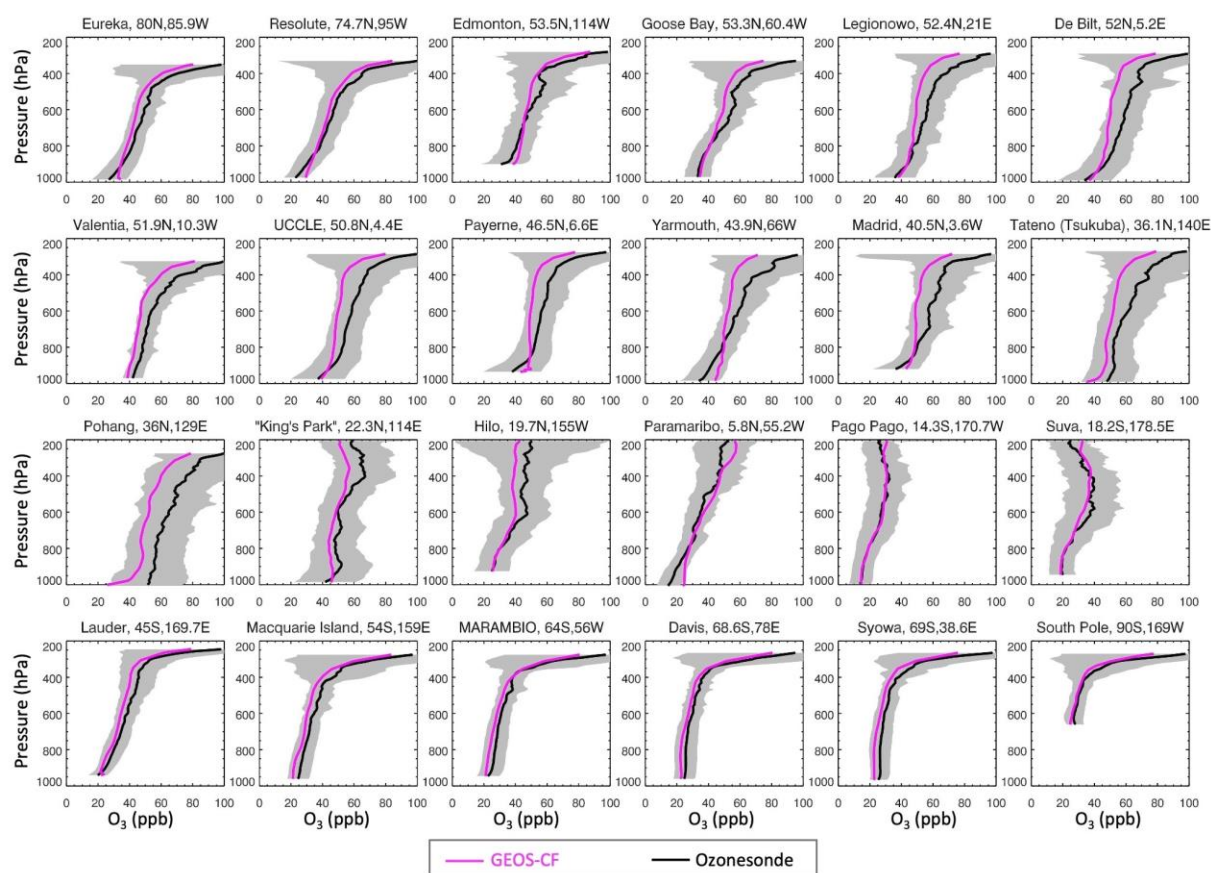399



**Figure 4.** Vertical profile of annual average ozone (2018-2019) at 24 sites as observed by
ozonesondes (black) and simulated by GEOS-CF (magenta). The grey shaded area indicates
sonde observation standard deviation.

405        The GAW observations show a pronounced seasonal cycle for surface $O_3$ with a
406    maximum of 30-50 ppbv during spring and summer and a low of 10-30 ppbv in winter, a feature
407    that is well reproduced by GEOS-CF (Figure 5). At remote locations such as the high-latitude
408    GAW sites (Figure 5 top row), the seasonal cycle is more distinct with a peak in early spring.
409    The ozone maximum develops more slowly at locations that are more heavily influenced by
410    anthropogenic emissions. As already observed in the ozonesondes, the model underestimates

411 ozone over the Southern Ocean by up to 10 ppbv. This is likely due to excessive ozone
412 deposition over seawater (Pound et al., 2020), a problem since corrected in newer versions of
413 GEOS-Chem.
414
415        Figure 6 shows monthly average surface ozone by region, as captured by observations in
416 the OpenAQ database and simulated by GEOS-CF. The model generally overestimates surface
417 $O_3$ over Europe, North and South America and Australia by 5-10 ppbv and underestimates it over
418 Africa and Asia by up to 20 ppbv. The model-observation mismatch is larger than for the remote
419 GAW sites, suggesting that sub-grid factors such as vertical and horizontal model representation
420 errors and nighttime loss of ozone by reaction with NO contribute to this difference (Travis et al.,
421 2019; Dacic et al., 2020). Models generally underestimate ozone nighttime depletion (Travis et
422 al., 2017) and evaluation of surface ozone is thus often restricted to daytime ozone (Hu et al.,
423 2018, Travis et al., 2019). As shown in Figure 7, restricting the analysis to daytime ozone (12:00
424 to 16:00 local time) does indeed improve the comparison and reduce the bias by up to 5 ppbv, in
425 particular over Europe, South America and Australia. The model still shows a systematic
426 positive bias over the US during summer and fall, a known issue in GEOS-Chem (Travis et al.,
427 2017, Hu et al., 2018).
428 Factors that likely contribute to the high NH surface ozone in GEOS-CF are uncertainties in the
429 production of ozone from the oxidation of isoprene (Travis et al., 2016; Bates et al., 2019) and
430 errors in ozone deposition to wet surfaces (Travis et al., 2019). In addition, most OpenAQ
431 observation sites are located near densely populated areas, and the 25x25 $km^2$ model simulation
432 cannot fully capture the fine-scale features characteristic for these environments (Keller et al.,
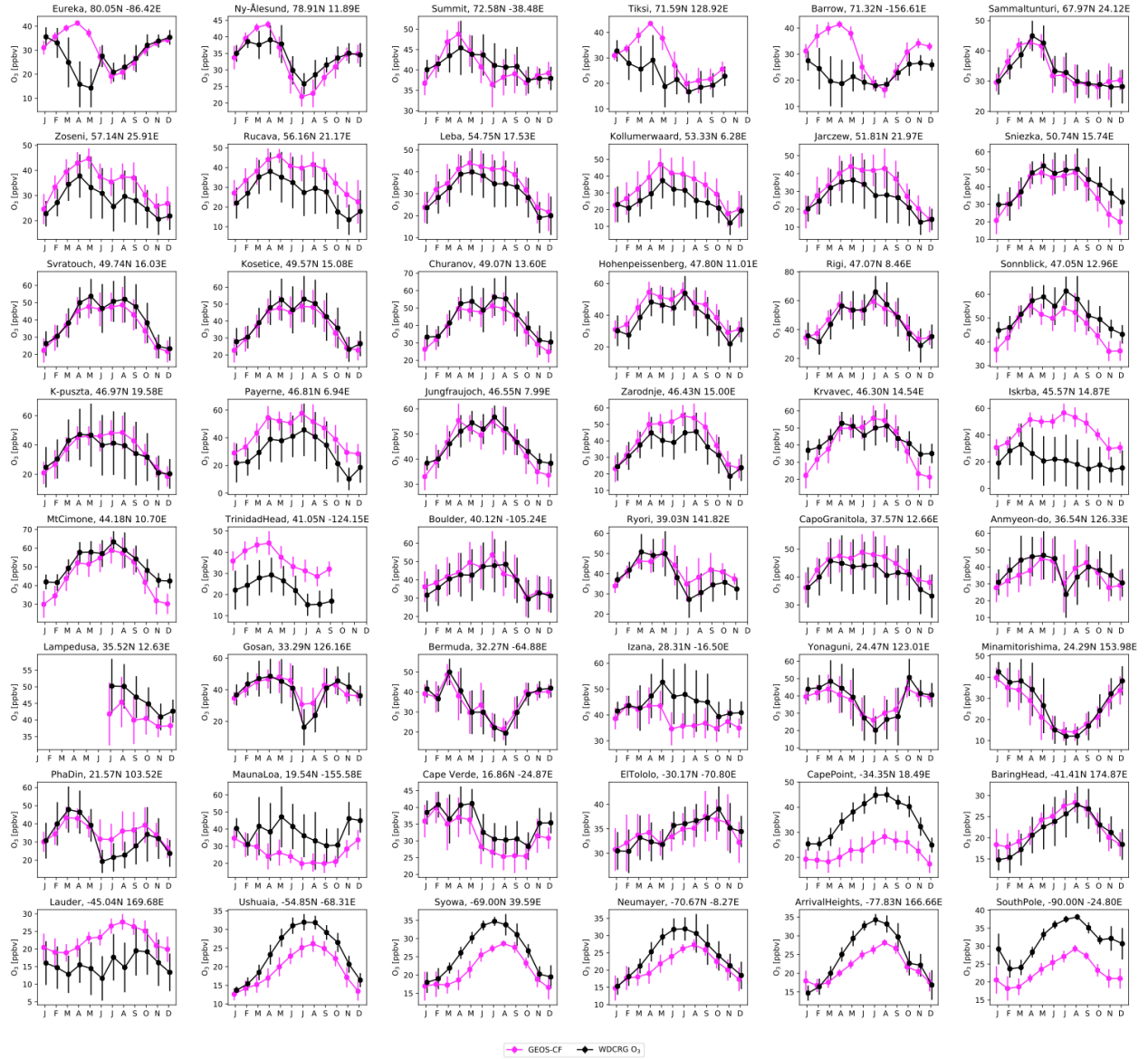433 2020).

**Figure 5.** Monthly average surface O$_3$ as observed at 48 GAW sites (black) and simulated by GEOS-CF (magenta). Vertical bars represent the standard deviation of hourly variability. Y-axis ranges vary by station.
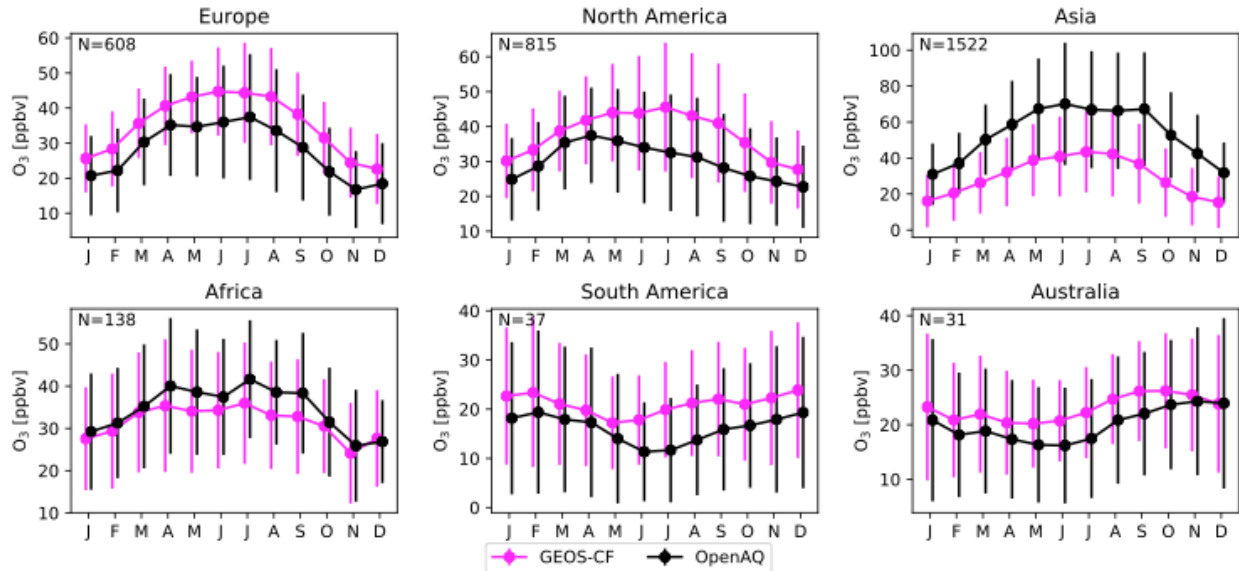
**Figure 6.** Monthly average surface $O_3$ grouped into six regions as obtained from the OpenAQ database (black) and simulated by GEOS-CF (magenta). Vertical bars represent the standard deviation of hourly variability across all sites. Number of sites is given in the inset. Y-axis ranges vary by region.



**Figure 7.** As Figure 6 but using afternoon ozone only (12:00-16:00 local time).

### 4.3. Nitrogen Dioxide

As shown in Figure 8, GEOS-CF captures major features of the global distribution of tropospheric $NO_2$. The model simulated $NO_2$ columns (Figure 8a) show distinct hot spots over urban areas (Eastern China, Europe, Eastern US), reflecting the dominant contribution of fossil fuel combustion coupled with the short atmospheric lifetime of $NO_2$ (Streets et al., 2013, Duncan et al., 2016). Additional hot spots resulting from biomass burning are found over Africa and the boreal region of Canada (Figure 8a). The spatial pattern simulated by GEOS-CF agrees well with the $NO_2$ columns observed by OMI. Over Eastern China, the model simulated $NO_2$ columns are

454  up to $3.0 \times 10^{15}$ molecules cm$^{-2}$ (or approx. 40%) higher than the OMI observations (Figure 8b),
455  suggesting a potential overestimation of NO$_x$ emissions or a longer NO$_x$ lifetime in the model
456  (Shah et al., 2020). However, the OMI retrieval algorithm v4.0 tends to underestimate
457  tropospheric NO$_2$ over polluted areas (Lamsal et al., 2020), which complicates the analysis. As
458  further discussed in the next section, the comparison against surface observations does not
459  support the view of a broad-based overestimation of surface NO$_2$ over Asia.

460  As shown in Figure 8c, the simulated seasonality of tropospheric NO$_2$ columns is in good
461  agreement with OMI observations. Over areas dominated by anthropogenic activities, such as the
462  US, Europe, and China, the simulated NO$_2$ columns show a distinct seasonal cycle with the
463  minimum during summer and peak during winter, driven by the seasonal variation in NO$_x$
464  lifetime against oxidation (Shah et al., 2020). The seasonal cycle observed over China is well
465  captured by GEOS-CF, while the simulated wintertime peak over the US and Europe is higher
466  than observed by OMI. Chemical loss of NO$_x$ during winter is dominated by N$_2$O$_5$ hydrolysis,
467  which is sensitive to NO$_x$ emissions and ozone concentrations (Jaeglé et al, 2018; Shah et al.,
468  2020). Over Africa, the seasonal cycle is dominated by summer biomass burning, which is well
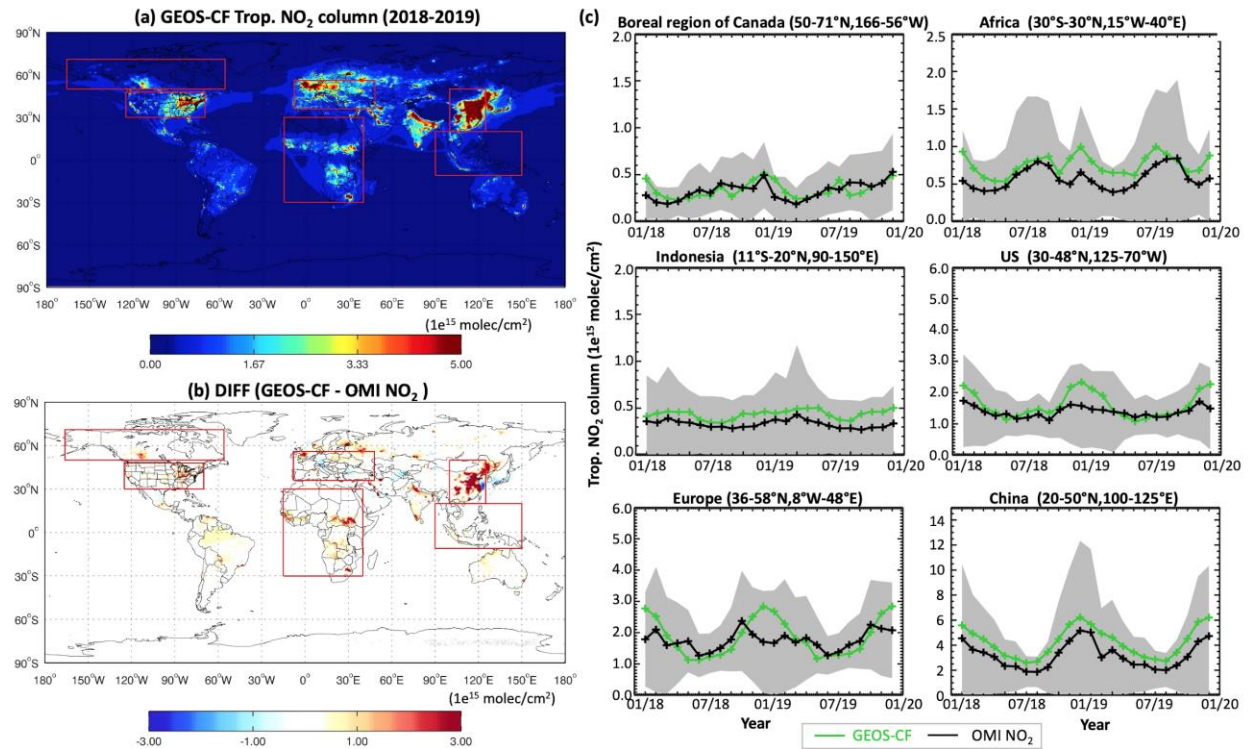469  captured by the model.

470



472  **Figure 8.** Comparison of GEOS-CF against OMI tropospheric column NO$_2$. **a)** the 2018-2019
473  average tropospheric NO$_2$ column (1-2pm local time) as simulated by GEOS-CF. **b)** the
474  difference between model simulated tropospheric column and NASA OMI NO$_2$ (v4.0)
475  observations. **c)** Right panel shows the tropospheric NO$_2$ time series averaged for six regions, as
476  shown in **a** and **b** (n.b., y-axis intervals are not the same for each time-series).

477

478  Figure 9 shows comparisons of monthly mean surface NO$_2$ at 6 GAW sites (n.b., all
479  located in Europe) against model simulated concentrations. These comparisons show that GEOS-
480  CF is in good agreement with the (non-mountainous) GAW sites, suggesting that it provides a

481    realistic simulation of background surface $NO_2$ over Europe. Figure 10 further evaluates the
482    simulated distribution of global surface $NO_2$ in comparison to observations in OpenAQ. The
483    model is in excellent agreement with observations over North America and Africa but
484    underestimates concentrations over Europe and South America, as well as Asia during the
485    wintertime. The apparent low bias over Europe and Asia is inconsistent with the tropospheric
486    column comparisons shown in Figure 8 and also the comparison against the GAW observations
487    (Figure 9), which do not show such a systematic underestimation of $NO_2$ by GEOS-CF.

488    The comparison of surface concentrations of $NO_2$ is complicated by the fact that most surface
489    observations are based on chemiluminescence using a molybdenum converter, which can result
490    in an overestimation of reported $NO_2$ concentrations due to interference with other oxidized
491    nitrogen compounds (Winer et al., 1974; Grosjean and Harrison, 1985; Steinbacher et al., 2007).
492    This might explain some of the model underestimation of $NO_2$ relative to the OpenAQ
493    observations. In addition, since the OpenAQ observations tend to be located in relative proximity
494    to urban areas they often do not represent the regional average $NO_2$ concentrations captured by
495    GEOS-CF, which can introduce a systematic bias. While this is an issue for all analyzed species,
496    the problem is particularly pronounced for $NO_2$ given its large spatial and temporal variability.
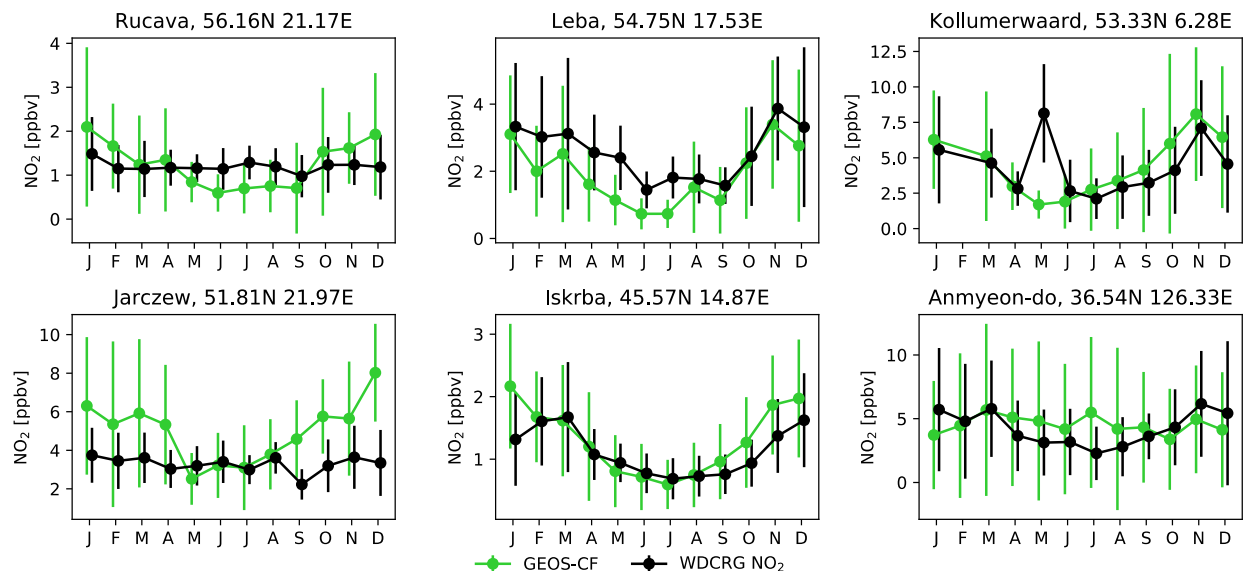
497



498
499    **Figure 9.** Monthly average surface $NO_2$ as observed at 6 GAW sites (black) and simulated by
500    GEOS-CF (green). Vertical bars represent the standard deviation of hourly variability. Y-axis
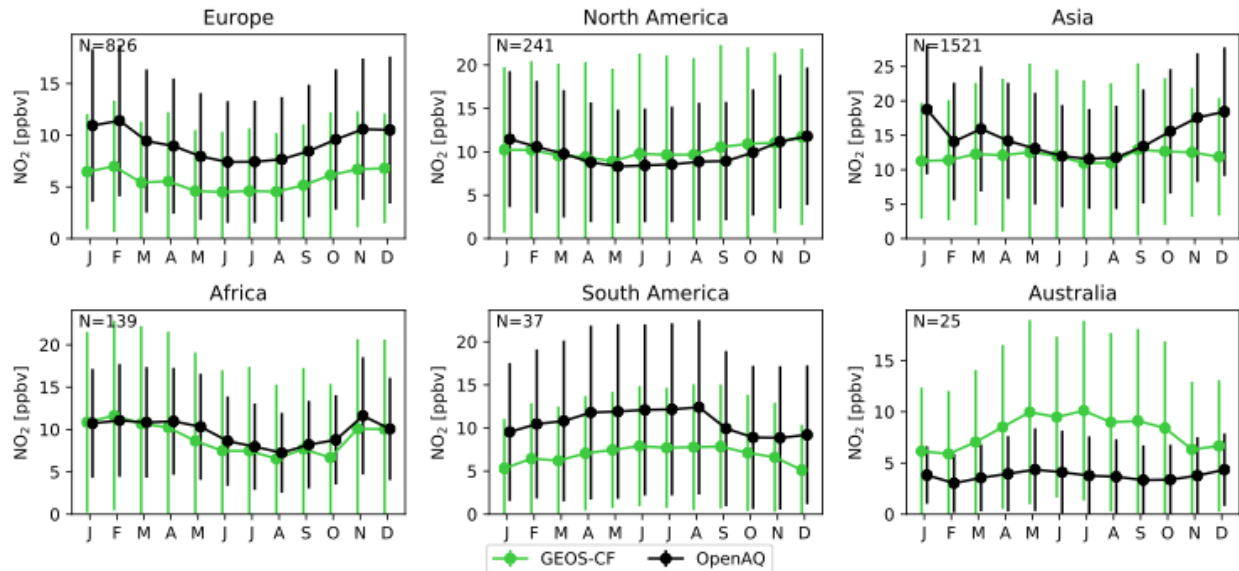501    ranges vary by station.

**Figure 10.** Monthly average surface $NO_2$ grouped into six regions as obtained from the OpenAQ database (black) and simulated by GEOS-CF (green). Vertical bars represent the standard deviation of hourly variability across all sites. Number of sites is given in the inset. Y-axis ranges vary by region.

## 4.4. Carbon Monoxide

Our analysis of simulated CO shows that the model has no significant global CO bias compared against satellite and surface observations. Figure 11 shows the global distribution and seasonal cycle of total column CO retrieved from MOPITT and derived from GEOS-CF. Local MOPITT averaging kernels were applied to the GEOS-CF CO profiles to obtain the column values. The model simulated CO pattern is in good agreement with MOPITT, with local maxima over major polluted areas (East China, India) and over biomass burning regions (South America and Central Africa). Similar to $NO_2$, the simulated CO columns over China are larger than the observations, possibly due to an overestimation of CO emissions over that region. The baseline anthropogenic CO emissions in GEOS-CF are scaled based on ODIAC emission trends, with strongest increases over China and India (Oda et al., 2018). Our scaling methodology assumes a constant $CO/CO_2$ emissions ratio, and any decrease in the $CO/CO_2$ emission ratio, e.g. due to a technology shift, would result in an overestimation of CO emissions.

The seasonal cycle of CO is determined by its shorter chemical lifetime during summer due to increased photochemical activity, which is reflected in lower NH concentrations during July and August where the model underestimates the MOPITT concentrations in the NH middle and high latitudes by 10-20% (Figure 11c). This underestimation is driven by a stronger than observed decrease of simulated total column CO during summertime, a pattern that is confirmed by comparisons against the GAW surface observations (Figure 12). While the model underestimates summertime surface CO in the NH, the opposite is true for the SH where model simulated concentrations during summer are higher than the observations. A low bias in CO is a long-standing issue in atmospheric chemistry models, commonly attributed to an underestimation of direct emissions and inconsistencies in the simulated distribution of the hydroxyl radical OH, the main atmospheric oxidant (Shindell et al., 2006; Strode et al., 2015, Flemming et al., 2015; Monks et al., 2015; Gaubert et al., 2016). The air mass-weighted mean

tropospheric OH in GEOS-CF is 11.9 x $10^5$ molecules cm$^{-3}$, in good agreement with other models as well as estimates derived from methyl chloroform observations (Spivakovsky et al., 2000; Montzka et al., 2000; Naik et al., 2013). The inter-hemispheric (NH/SH) ratio is 1.33, again in good agreement with other model estimates (Naik et al., 2013) but higher than observation-derived values of 0.8-1.0 (Montzka et al., 2000; Prinn et al. 2001; Krol and Lelieveld 2003; Patra et al., 2014). This is consistent with an underestimation of summertime CO in the northern high-latitudes - as shown by the comparisons against MOPITT and the GAW surface observations (Figures 11 and 12) - and an overestimation of SH CO (Figure 12). An overestimation of NH OH in GEOS-CF is also supported by inversion studies using the GEOS-Chem CTM, which find that a similar OH correction is needed to match methane observations (Zhang et al., 2020).
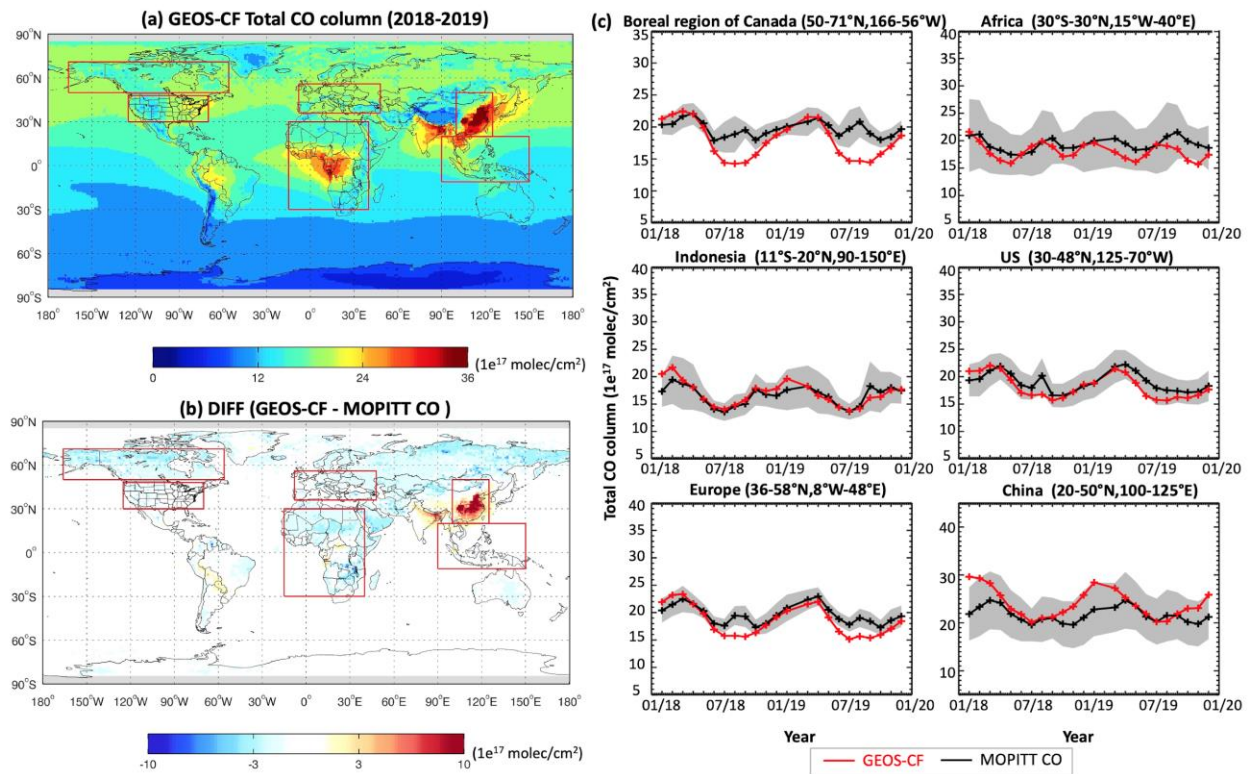


**Figure 11.** Comparison of GEOS-CF against MOPITT V8 total column CO. Top left panel shows the 2018-2019 average total CO column as simulated by GEOS-CF. Bottom left panel shows the difference between model simulated total column and MOPITT observations. Right panel shows the total CO time series averaged for six regions, as shown on the left.
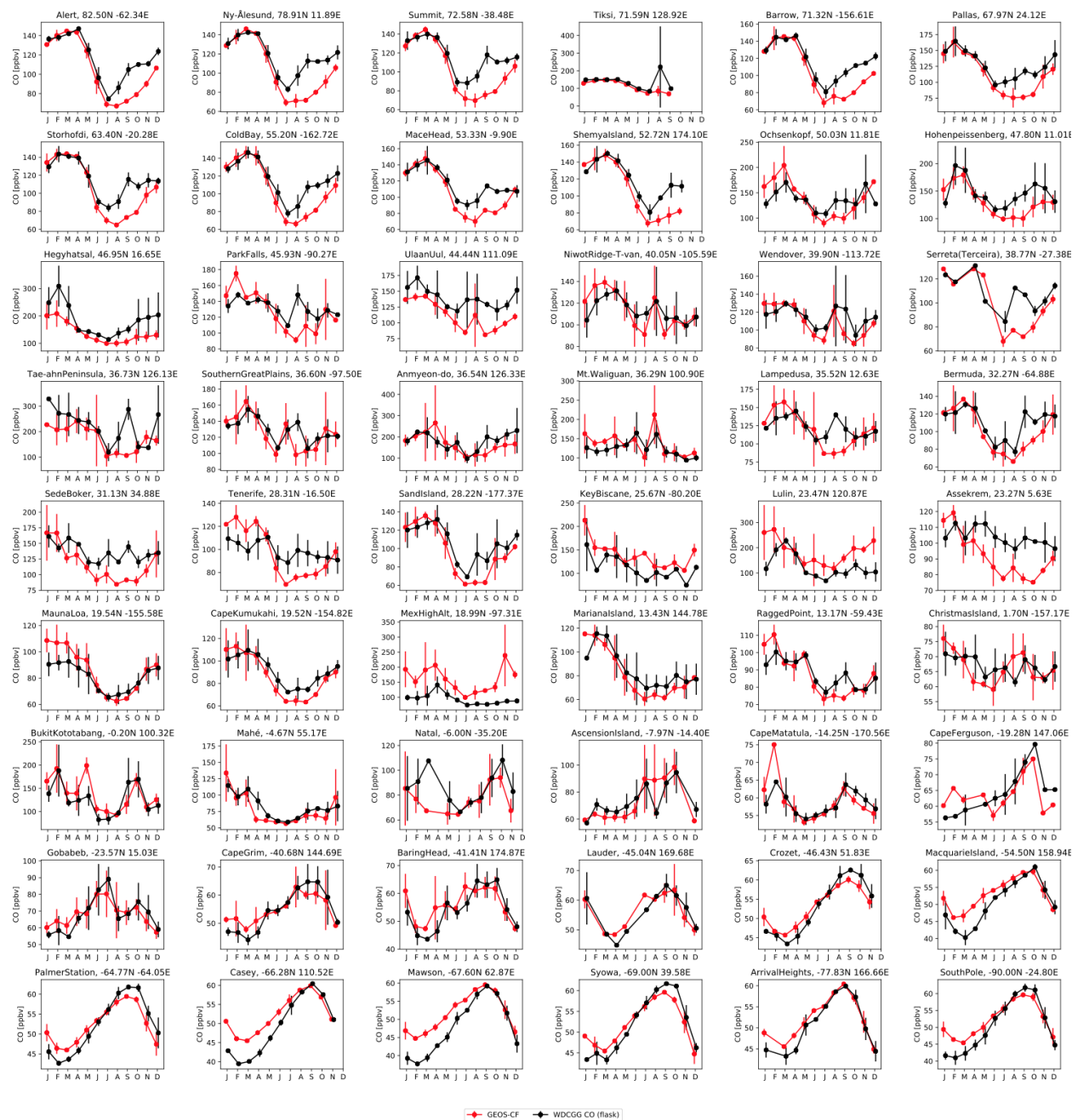
549
550 **Figure 12.** Monthly average surface CO as observed at 54 GAW sites (black) and simulated by
551 GEOS-CF (red). Vertical bars represent the standard deviation of hourly variability. Y-axis
552 ranges vary by station.

553        4.5. Aerosols

554        The evaluation of model simulated $O_3$, $NO_2$ and CO has shown that GEOS-CF
555 reproduces many of the features of the tropospheric distribution of these trace gases. With
556 respect to aerosols simulated by GEOS-Chem, our validation shows that GEOS-CF has a high
557 bias but still captures many of the observed spatial and temporal patterns. A high bias in aerosols
558 is a known issue in GEOS-Chem v12.0.1 used in GEOS-CF v1.0, and recent versions of GEOS-

559  Chem show improved simulation of aerosols including surface PM$_{2.5}$ (Luo et al., 2019; 2020;
560  Carter et al., 2020).

561       Figure 13 compares model simulated AOD at 550nm against AERONET observations,
562  and Figure 14 shows corresponding model-observation comparisons for surface PM$_{2.5}$. GEOS-
563  CF overestimates the observations for both AOD and surface PM$_{2.5}$, pointing toward a systematic
564  overestimation of aerosols in the model. On a relative basis, the overprediction is most
565  pronounced for Europe and North America, and the largest absolute bias of surface PM$_{2.5}$ is
566  found over Asia. We attribute part of the model overestimation to inadequate treatment of wet
567  scavenging processes, which results in an overprediction of aerosol nitrate and ammonium,
568  especially over Asia (Luo et al., 2019; 2020). Further, the QFED biomass burning emissions
569  inventory uses enhancement factors based on the GOCART model to better match MODIS-
570  observed AOD (Darmenov and Da Silva, 2015), which results in an overestimation of biomass
571  burning emissions if the differences between the GEOS-Chem and GOCART aerosol
572  parameterizations are not accounted for (Carter et al., 2020).
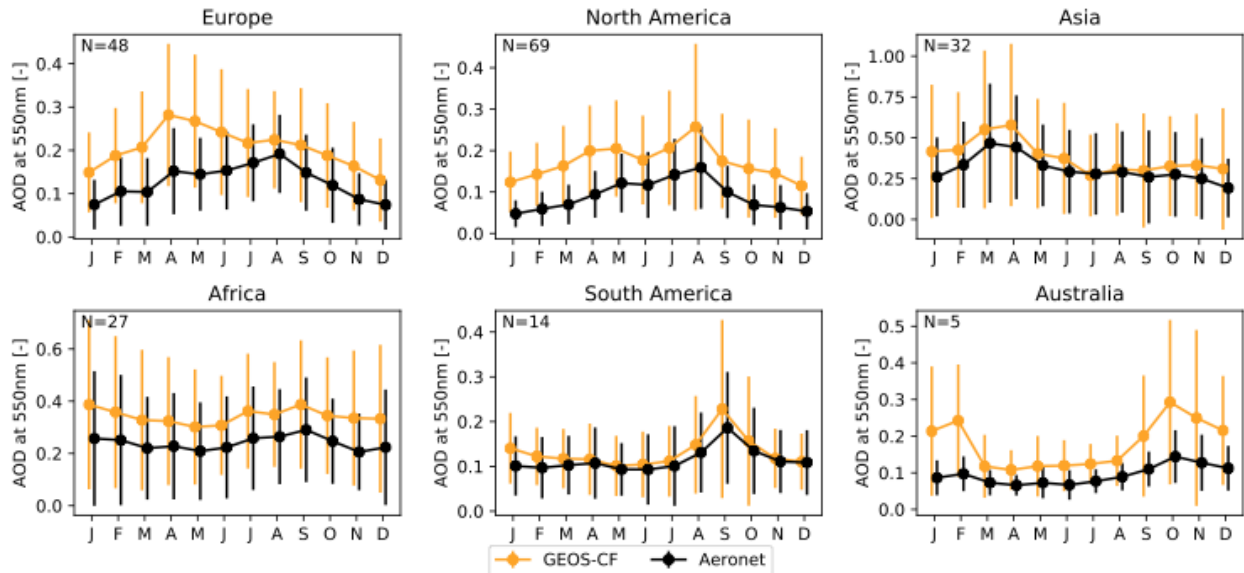
573

574



575  **Figure 13.** Monthly average Aerosol optical depth (AOD) at 550nm grouped into six regions as
576  observed at AERONET sites (black) and simulated by GEOS-CF (orange). Vertical bars
577  represent the standard deviation of daily variability. Number of sites is given in the inset. Y-axis
578  ranges vary by region.

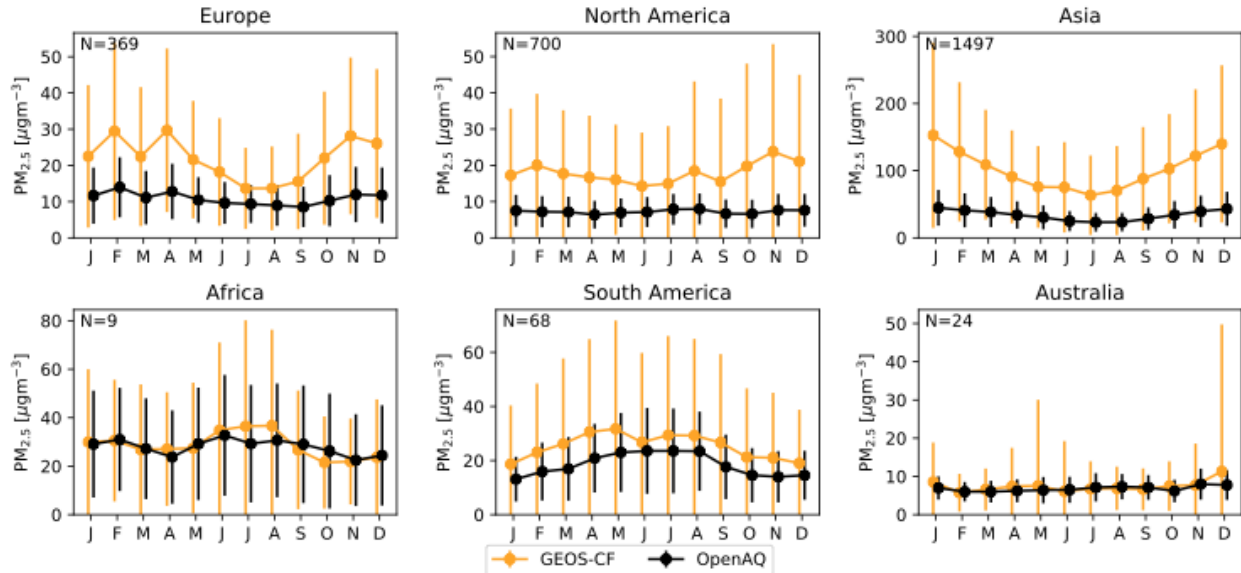**Figure 14:** Monthly average surface PM$_{2.5}$ grouped into six regions as obtained from the OpenAQ database (black) and simulated by GEOS-CF (orange). Vertical bars represent the standard deviation of hourly variability. Number of sites is given in the inset. Y-axis ranges vary by region.

## 5 Evaluation of Model Forecasts

### 5.1. Comparison of Model Forecasts against Observations

GEOS-CF v1.0 does not directly assimilate trace gas observations and differences between the 1-day analysis and the model forecasts are thus mainly driven by variations between the forecasted and analyzed meteorological state. The meteorology not only impacts the flow of the constituents but also affects deposition and dynamically calculated emissions, including lightning NO$_x$, biogenic VOCs, sea salt aerosols, and dust (see Table 1). Further, the model forecasts assume persistence in the biomass burning emissions, meaning the fires observed during the analysis are assumed to continue burning and emitting the same amount for the next five days. The model thus does not capture changes in occurrence or intensity of wildfires, both of which can lead to significant changes in surface air pollution close to and downwind from the fires.

Figure 15 shows model-observation skill scores for the model analysis (forecast day -1) and the 5-day forecasts (forecast days +1 to +5) for O$_3$, NO$_2$, and PM$_{2.5}$. The results for the analysis are the same as discussed in Section 4 (Figure 3). Skill scores were calculated at each observation site individually before aggregating them in the form of boxplots, as shown in Figure 15. For all three evaluated species, the model analysis showed the best agreement with the observations. The median NMB of the model analysis is -0.01 for O$_3$, -0.26 for NO$_2$, and 1.50 for PM$_{2.5}$, with almost no difference between the analysis and the model forecasts. Relative to the analysis, the forecasted NRMSE and R become incrementally worse for the 1-day to 3-day lead forecasts, while there is little further deterioration between the 3-day forecasts and the 5-day forecasts. The median NRMSE is 0.36 for O$_3$, 0.42 for NO$_2$ and 1.05 for PM$_{2.5}$. The NRMSE slightly deteriorates with increasing forecast lead time, resulting in NRMSE's for the 5-day forecast of 0.4 for O$_3$, 0.45 for NO$_2$ and 1.07 for PM$_{2.5}$. This indicates that errors in the

608  meteorological forecasts (and biomass burning emissions) indeed impact the quality of the
609  surface air quality forecasts. The increase in NRMSE is most pronounced for $O_3$, whose
610  chemistry is strongly controlled by meteorological factors such as solar radiation, temperature,
611  and humidity (e.g., Jacob et al., 1993; Sillman and Samson, 1995, Tarasova and Karpetchko,
612  2003). The largest change in skill score between analysis and forecasts is found for R, which
613  drops from 0.6 for the analysis to 0.44 for the 5-day forecast for $O_3$, 0.32 to 0.15 for $NO_2$, and
614  0.47 to 0.17 for $PM_{2.5}$. Of the three analyzed skill scores, R is most sensitive to errors in the
615  temporal pattern and we attribute a large fraction in the deterioration in R to transport errors in
616  the forecasts, such as the evolution of frontal systems or the dispersion of smoke plumes. $PM_{2.5}$
617  is particularly sensitive to these factors given its large spatiotemporal gradients, comparatively
618  long atmospheric lifetime, and strong sensitivity to changes in biomass burning emissions.
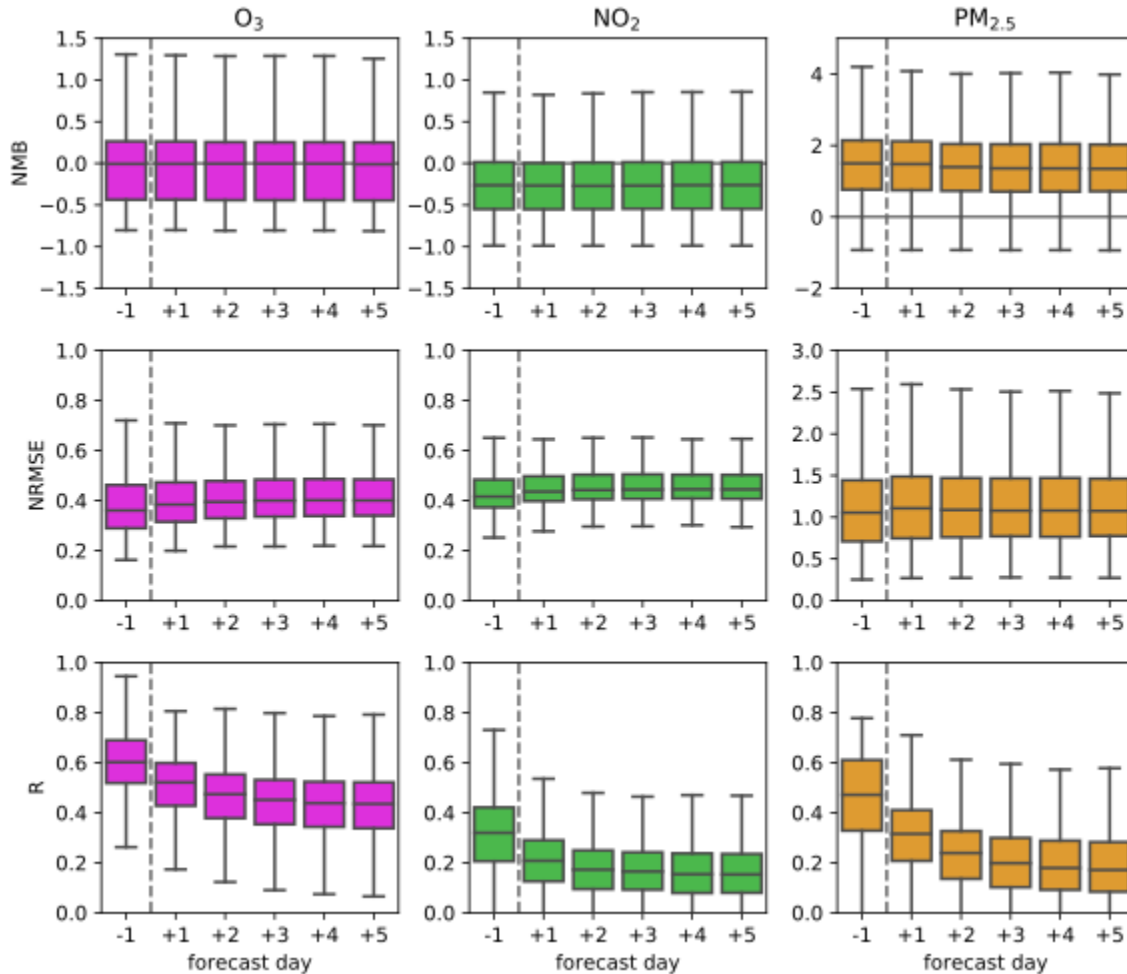


**Figure 15.** GEOS-CF model skill scores for the analysis (forecast day -1) and the 5-day forecasts
(forecast day +1 to +5) relative to surface observations. Boxplots show the variation in the NMB,
NRMSE, and R across all surface sites for $O_3$, $NO_2$, and $PM_{2.5}$.

### 5.2. Bias-corrected Local Forecasts

624  As discussed above, model-predicted concentrations of $O_3$, $NO_2$ and $PM_{2.5}$ can differ
625  from the observations for a number of reasons, including model representation errors,
626  uncertainties in the meteorology, or model biases arising from errors in the model treatment of

627 emissions, deposition, or atmospheric chemistry. One approach to deal with these issues is to
628 quantify and correct these systematic model errors in a post-processing step. Such bias correction
629 methods can be applied to near real-time model forecasts and have been found to be an effective
630 tool to significantly improve local model predictions, e.g., by using mean subtraction (McKeen
631 et al., 2005; Wilczak et al., 2006), historical analogs (Hamill and Whitaker, 2006), Kalman-
632 filtering (Delle Monache et al., 2006; Djalalova et al., 2015), or kriging (Honoré et al., 2008).
633 More recently, machine learning (ML) approaches have become popular to relate model output
634 to air quality observations (e.g., Grange et al., 2018; Grange and Carslaw, 2019; Ivatt and Evans,
635 2020; Petetin et al., 2020). As discussed in Keller et al. (2020), bias-correction using ML can
636 significantly reduce GEOS-CF model biases compared to surface observations. This is illustrated
637 in Figure 16, which shows the GEOS-CF model skill scores for the same data set analyzed in
638 Section 8 but using bias-corrected model concentrations instead of the original model output.
639 The bias correction methodology is described in detail in Keller et al. (2020). It uses the
640 XGBoost algorithm (Chen and Guestrin, 2016) to correct the original model predictions of $O_3$,
641 $NO_2$, and $PM_{2.5}$ based on local meteorology and composition, as predicted by the GEOS-CF
642 model. The ML algorithm was trained on analysis data for year 2018 and the results shown in
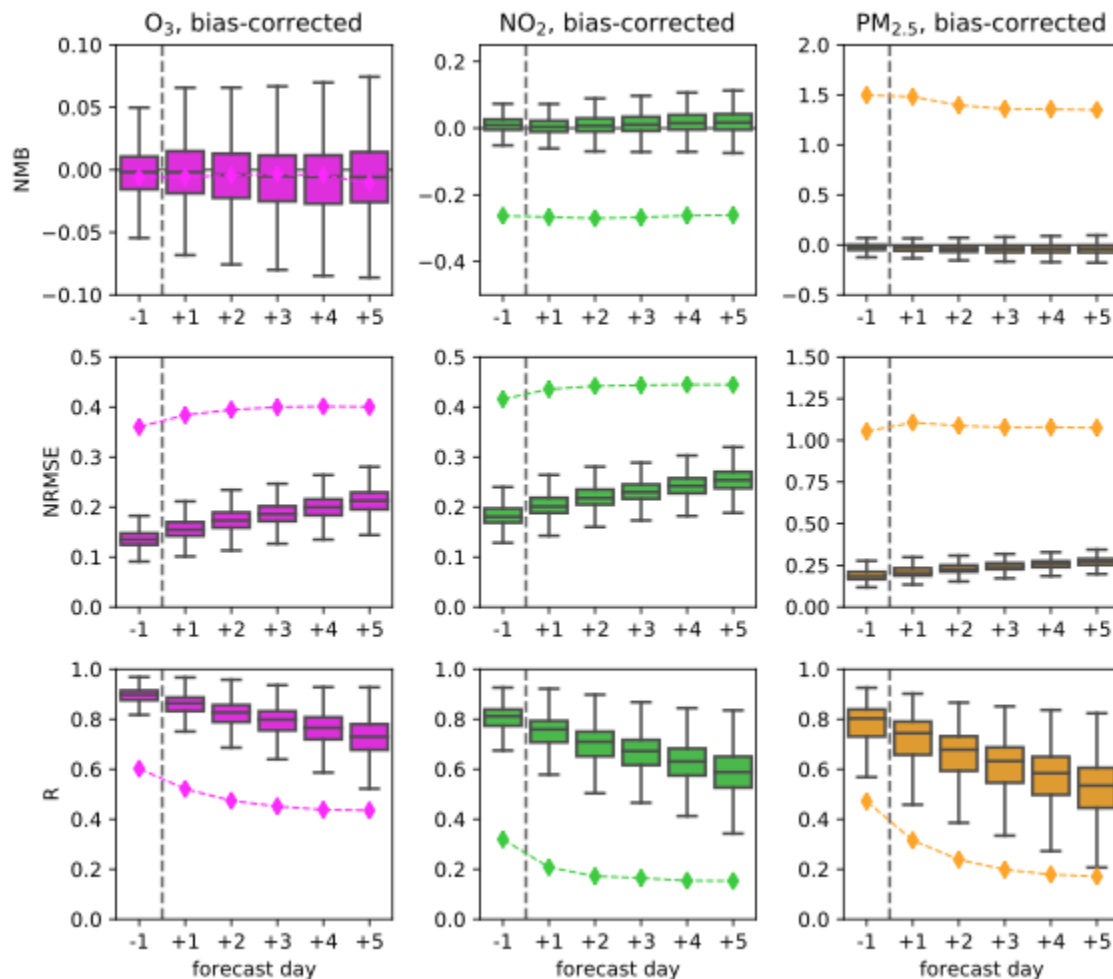643 Figure 16 are for year 2019.
644



645
646 **Figure 16.** Similar to Figure 14 but boxplots indicate the GEOS-CF model skill scores for the
647 bias-corrected analysis (forecast day -1) and the bias-corrected 5-day forecasts (forecast day +1

648 to +5) relative to surface observations as boxplots and the median statistics of the non-corrected
649 model values from Figure 15 shown as diamonds.

650

651 Compared to the uncorrected model output (indicated by the diamonds in Figure 16), the
652 bias-corrected model values agree much better with the observations for all species, skill scores,
653 and lead times. For all three species, the NMB of the bias-corrected values is close to zero for
654 both the analysis and the 5-day forecasts. This result is not unexpected given that the ML
655 algorithm is designed to minimize the model bias. The NRMSE and R scores are also
656 significantly improved compared to the original data, with RMSE values of 0.1-0.2 and R values
657 between 0.6-0.9 for the analysis. The skill scores of the bias-corrected forecasts deteriorate more
658 rapidly than is the case for the uncorrected output but still outperform the original forecasts for
659 all five lead days. Since the ML algorithm was only trained on the analysis data, model errors in
660 the meteorology forecasts will also negatively impact the quality of the bias correction applied to
661 the (forecast) baseline, which explains the steady decline in the skill scores for the bias-corrected
662 forecasts.

663 **6 Conclusions**

664 The GEOS-CF system provides global, near real-time and 5-day forecast simulations of
665 atmospheric composition and meteorology at the high global resolution of 0.25 degrees --
666 including 5-day forecasts -- based on the GEOS-Chem chemistry module online within the
667 GEOS GCM. Comparisons against a suite of satellite, ozonesonde and surface observations
668 demonstrate that the model realistically captures the global distribution of a wide range of air
669 constituents, including $O_3$, $NO_2$, and CO. For these three species, the model shows little biases at
670 background locations, with NMB values ranging between -0.1 to +0.1. Like other atmospheric
671 chemistry models, the model overestimates surface ozone in the NH during summer, especially
672 over the south eastern US (Emmons et al., 2020, Fleming et al., 2015, Travis et al., 2016). While
673 the horizontal resolution of 0.25 degrees is one of the highest for a global atmospheric chemistry
674 forecast model, it is still not fine enough to resolve some of the meso-scale features of urban air
675 pollution, which can lead to substantial model-observation mismatches when compared against
676 urban air quality observations. Applying a bias-correction algorithm to the gridded model output,
677 based on ML using historical observation-model comparisons, can lead to significant
678 improvements of the model predictions over urban areas. This procedure, which can be
679 automated as a post-processing step, offers an interesting option to provide highly localized
680 forecasts at selected locations.

681

682 The GEOS-CF modeling system leverages components developed by the GEOS and the
683 GEOS-Chem modeling communities and directly benefits from the continuous advancements
684 provided by these groups. The current version of GEOS-CF (v1.0) incorporates GEOS-Chem
685 v12.0.1. Several important updates have been added to GEOS-Chem since, and those will be
686 included in future versions of GEOS-CF. This includes updates to the chemistry of isoprene
687 (Bates and Jacob, 2019) and halogens (Wang et al., 2019), improved wet scavenging of aerosols
688 (Luo et al., 2019; 2020), and updated ozone deposition over seawater (Pound et al., 2020). When
689 implemented in future versions of GEOS-CF, these updates are expected to reduce the high bias
690 observed in $PM_{2.5}$ and AOD as well as the high bias in surface ozone over the southern
691 hemisphere.

Another model development focus will center around the assimilation of satellite observations of atmospheric constituents, which has been shown to lead to improved representation of atmospheric composition, in particular for longer-lived species such as $O_3$ and CO (Flemming et al., 2015). In its current form, GEOS-CF does not directly assimilate tropospheric trace gas observations, and the variability in constituent distribution is thus driven by the anthropogenic emission inventories, real-time biomass burning information, and the current meteorological state and its impact on dynamic emission sources such as biogenic emissions or sea salt aerosols. It should be noted that the anthropogenic emission inventories do not incorporate short-term, real-time changes in emissions, e.g., due to reduced human activities in the wake of the COVID-19 pandemic, and GEOS-CF v1.0 thus represents a business-as-usual estimate of the atmosphere (Keller et al., 2020). The inclusion of near real-time information for dynamic emission adjustment, e.g., based on traffic data or satellite observations, will be another model development focus.

GEOS-CF offers a new tool for academic researchers, air quality managers, and the public. Applications include flight campaign planning, support of satellite and other remote-sensing observations, interpretation of field campaign data (Dacic et al., 2020), and air quality research (Keller et al., 2020).

## Data Availability Statement

All model output is centrally stored at the NASA Center for Climate Simulation (NCCS). Public access to these archives is provided by the GMAO at https://gmao.gsfc.nasa.gov/weather_prediction/GEOS-CF/data_access/ in the form of weather maps and through model output access tools, including OPeNDAP and Hypertext Transfer Protocol (HTTP).

## References

Amos, H. M., D. J. Jacob, C. D. Holmes, J. A. Fisher, Q. Wang, R. M. Yantosca, E. S. Corbitt, E. Galarneau, A. P. Rutter, M. S. Gustin, A. Steffen, J. J. Schauer, J. A. Graydon, V. L. St. Louis, R. W. Talbot, E. S. Edgerton, Y. Zhang, and E. M. Sunderland (2012), Gas-Particle Partitioning of Atmospheric Hg(II) and Its Effect on Global Mercury Deposition, *Atmos. Chem. Phys.,*12,591-603.

Bacmeister, J. T., Suarez, M. J., & Robertson, F. R. (2006). Rain reevaporation, boundary layer convection interactions, and Pacific rainfall patterns in a AGCM. *Journal of the Atmospheric Sciences*, 63, 3383–3403. https://doi.org/10.1175/JAS3791.1

Bates, K.H., and D.J. Jacob (2019), A new model mechanism for atmospheric oxidation of isoprene: global effects on oxidants, nitrogen oxides, organic products, and secondary organic aerosol, *Atmos. Chem. Phys.*, 19, 9613-9640.

Bey, I., Jacob, D.J., Yantosca, R.M., Logan, J.A., Field, B.D., Fiore, A.M., Li, Q., Liu, H.Y., Mickley, L.J., Schultz, M.G. (2001), Global modeling of tropospheric chemistry with assimilated

736    meteorology: Model description and evaluation. *J. Geophys. Res. Atmospheres* 106, 23073–
737    23095. https://doi.org/10.1029/2001JD000807

738    Bhattacharjee, P. S., Wang, J., Lu, C.-H., and Tallapragada, V. (2018), The implementation of
739    NEMS GFS Aerosol Component (NGAC) Version 2.0 for global multispecies forecasting at
740    NOAA/NCEP – Part 2: Evaluation of aerosol optical thickness, *Geosci. Model Dev.,* 11, 2333–
741    2351, https://doi.org/10.5194/gmd-11-2333-2018.

742    Bian, H., Prather, M.J. (2002), Fast-J2: Accurate Simulation of Stratospheric Photolysis in
743    Global Chemical Models. *J. Atmospheric Chem.* 41, 281–296.
744    https://doi.org/10.1023/A:1014980619462

745    Borovikov, A., Cullather, R., Kovach, R. et al. (2019), GEOS-5 seasonal forecast system. *Clim*
746    *Dyn 53*, 7335–7361. https://doi.org/10.1007/s00382-017-3835-2

747    Buchard, V., Randles, C.A., Silva, A.M. da, Darmenov, A., Colarco, P.R., Govindaraju, R.,
748    Ferrare, R., Hair, J., Beyersdorf, A.J., Ziemba, L.D., Yu, H., (2017), The MERRA-2 Aerosol
749    Reanalysis, 1980 Onward. Part II: Evaluation and Case Studies. *J. Clim. 30*, 6851–6872.

750    Carn, S., (2019), Multi-Satellite Volcanic Sulfur Dioxide L4 Long-Term Global Database V3.
751    https://doi.org/10.5067/measures/so2/data404

752    Carter, T. S., Heald, C. L., Jimenez, J. L., Campuzano-Jost, P., Kondo, Y., Moteki, N., Schwarz,
753    J. P., Wiedinmyer, C., Darmenov, A. S., da Silva, A. M., and Kaiser, J. W. (2020), How
754    emissions uncertainty influences the distribution and radiative impacts of smoke from fires in
755    North America, *Atmos. Chem. Phys.,* 20, 2073–2097, https://doi.org/10.5194/acp-20-2073-2020.

756    Chen, T. and C. Guestrin (2016), XGBoost: A Scalable Tree Boosting System, *CoRR*,
757    abs/1603.02754, 785–794, https://doi.org/10.1145/2939672.2939785.

758    Chou, M.-D. (1990). Parameterizations for the absorption of solar radiation by $O_2$ and $CO_2$ with
759    applications to climate studies. *Journal of Climate*,3, 209–217.

760    Chou, M.-D. (1992). A solar radiation model for use in climate studies. *Journal of the*
761    *Atmospheric Sciences*, 49, 762–772.

762    Chou, M.-D., & Suarez, M. J. (1994). An efficient thermal infrared radiation parameterization
763    for use in general circulation models. *NASA Tech.Memorandum*, NASA/TM-1994–104606, Vol.
764    3, 85 p.. Greenbelt, MD: NASA Goddard Space Flight Center.

765    Colarco, P., da Silva, A., Chin, M., Diehl, T. (2010). Online simulations of global aerosol
766    distributions in the NASA GEOS-4 model and comparisons to satellite and ground-based aerosol
767    optical depth. *J. Geophys. Res. Atmospheres* 115, D14207,
768    https://doi.org/10.1029/2009JD012820.

769    Croft, B., Wentworth, G., Martin, R. et al. (2016), Contribution of Arctic seabird-colony
770    ammonia to atmospheric particles and cloud-albedo radiative effect. *Nat Commun* 7, 13444.
771    https://doi.org/10.1038/ncomms13444

772    Dacic, N., John T. Sullivan, K. Emma Knowland, Glenn M. Wolfe, Luke D. Oman, Timothy A.
773    Berkoff, Guillaume P. Gronoff (2020), Evaluation of NASA's high-resolution global
774    composition simulations: Understanding a pollution event in the Chesapeake Bay during the
775    summer 2017 OWLETS campaign, *Atmospheric Environment,* Volume 222, 117133, ISSN
776    1352-2310, https://doi.org/10.1016/j.atmosenv.2019.117133.

Darmenov, A.S., da Silva, A. (2015). The Quick Fire Emissions Dataset (QFED)— Documentation of versions 2.1, 2.2 and 2.4. *Technical Report Series on Global Modeling and Data Assimilation*. NASA//TM-2015-104606, Vol. 38, 212 pp.

Deeter, M. N., Edwards, D. P., Francis, G. L., Gille, J. C., Mao, D., Martinez-Alonso, S., Worden, H. M., Ziskin, D., and Andreae, M. O. (2019), Radiance-based retrieval bias mitigation for the MOPITT instrument: the version 8 product, *Atmospheric Measurement Techniques*, 12, 4561-4580, 10.5194/amt-12-4561-2019.

Delle Monache, L., T. Nipen, X. Deng, Y. Zhou, and R. Stull (2006), Ozone ensemble forecasts: 2. A Kalman filter predictor bias correction, *J. Geophys. Res.*, 111, p. D05308, 10.1029/2005JD006311

Delle Monache, L., Wilczak, J., Mckeen, S., Grell, G., Pagowski, M., Peckham, S., Stull, R., Mchenry, J. & Mcqueen, J. (2008) A Kalman-filter bias correction method applied to deterministic, ensemble averaged and probabilistic forecasts of surface ozone, *Tellus B: Chemical and Physical Meteorology*, 60:2, 238-249, DOI: 10.1111/j.1600-0889.2007.00332.x

Djalalova, I., Delle Monache, L., and Wilczak J. (2015), PM2.5 analog forecast and Kalman filter post-processing for the Community Multiscale Air Quality (CMAQ) model, *Atmospheric Environment*, Volume 108, Pages 76-87, ISSN 1352-2310, https://doi.org/10.1016/j.atmosenv.2015.02.021.

Dlugokencky, E. J., L. P. Steele, P. M. Lang, and K. A. Masarie (1994), The growth rate and distribution of atmospheric methane, *J. Geophys. Res*., 99, 17,021– 17,043, doi:10.1029/94JD01245.

Douglass, A. R., Stolarski, R. S., Strahan, S. E., & Connell, P. S. (2004). Radicals and reservoirs in the GMI chemistry and transport model: Comparison to measurements. *Journal of Geophysical Research*, 109, D16302. https://doi.org/10.1029/2004JD004632

Douglass, A. R., Strahan, S. E., Oman, L. D., & Stolarski, R. S. (2014). Understanding differences in chemistry climate model projections of stratospheric ozone. *Journal of Geophysical Research: Atmospheres*, 119, 4922–4939. https://doi.org/10.1002/2013JD021159

Duncan, B. N., Logan, J. A., Bey, I., Megretskaia, I. A., Yantosca, R. M., Novelli, P. C., Rinsland, C. P. (2007). The global budget of CO, 1988– 1997: Source estimates and validation with a global model. *Journal of Geophysical Research,* 112, D22301. https://doi.org/10.1029/2007JD008459

Duncan, B. N. , L. N. Lamsal, A. M. Thompson, Y. Yoshida, Z. Lu, D. G. Streets, M. M. Hurwitz, and K. E. Pickering (2016), A space-based, high-resolution view of notable changes in urban NOx pollution around the world (2005–2014). *J. Geophys. Res.* 121, 976–996.

Eastham, S.D., Weisenstein, D.K., Barrett, S.R.H., (2014), Development and evaluation of the unified tropospheric–stratospheric chemistry extension (UCX) for the global chemistry- 34 transport model GEOS-Chem. *Atmos. Environ.* 89, 52–63.

Emmons, L. K., Schwantes, R. H., Orlando, J. J., Tyndall, G., Kinnison, D., Lamarque, J.-F., et al. (2020). The Chemistry Mechanism in the Community Earth System Model version 2 (CESM2). *Journal of Advances in Modeling Earth Systems*, 12, e2019MS001882. https://doi.org/10.1029/2019MS001882

818     Evans, M. J., and Jacob, D. (2005). Impact of new laboratory studies of $N_2O_5$ hydrolysis on
819     global model budgets of tropospheric nitrogen oxides, ozone, and OH, *Geophys. Res. Lett.,* 32,
820     http://doi/.org/10.1029/2005gl022469

821     Eyring, V., T. G. Shepherd, & D. W. Waugh (Eds.) (2010). SPARC report on the evaluation of
822     chemistry-climate models (SPARC Rep. 5, WCRP-132, WMO/TD 1526). Retrieved from
823     http://www.atmosp.physics.utoronto.ca/SPARC/ccmval_final

824     Frith, S. M., N. A. Kramarova, R. S. Stolarski, R. D. McPeters, P. K. Bhartia, and G. J. Labow
825     (2014), Recent changes in total column ozone based on the SBUV Version 8.6 Merged Ozone
826     Data Set, *J. Geophys. Res. Atmos.*, 119, 9735–9751, doi:10.1002/2014JD021889.

827     Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M.,
828     Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E.,
829     Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A. (2015).
830     Tropospheric chemistry in the Integrated Forecasting System of ECMWF, *Geosci. Model Dev.*,
831     8, 975–1003, https://doi.org/10.5194/gmd-8-975-2015.

832     Fountoukis, C., Nenes, A. (2007). ISORROPIA II: a computationally efficient thermodynamic
833     equilibrium model for K+–Ca2+–Mg2+–NH4 +–Na+–SO4 2 –NO3 - –Cl- –H2O aerosols.
834     *Atmospheric Chem. Phys.* 7, 4639–4659

835     Garcia, R. R., & Boville, B. A. (1994). Downward control of the mean meridional circulation
836     and temperature distribution of the polar winter stratosphere. *Journal of the Atmospheric*
837     *Sciences*, 51, 2238–2245.

838     Gaubert, B., et al. (2016), Toward a chemical reanalysis in a coupled chemistry-climate model:
839     An evaluation of MOPITT CO assimilation and its impact on tropospheric composition, *J.*
840     *Geophys. Res. Atmos.,* 121, 7310–7343, doi:10.1002/2016JD024863.

841     Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A.,
842     Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C.,
843     Akella, S., Buchard, V., Conaty, A., da Silva, A. M., Gu, W., Kim, G.- K., Koster, R., Lucchesi,
844     R., Merkova, D., Nielsen, J. E., Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert,
845     S. D., Sienkiewicz, M., and Zhao, B. (2017). The Modern-Era Retrospective Analysis for
846     Research and Applications, Version 2 (MERRA-2), *J. Climate,* 30, 5419–5454,
847     https://doi.org/10.1175/JCLI-D-16- 0758.1.

848     Giles, D. M., Sinyuk, A., Sorokin, M. G., Schafer, J. S., Smirnov, A., Slutsker, I., et al. (2019).
849     Advancements in the Aerosol Robotic Network (AERONET) Version 3 database—Automated
850     near-real-time quality control algorithm with improved cloud screening for Sun photometer
851     aerosol optical depth (AOD) measurements. *Atmospheric Measurement Techniques*, 12, 169–
852     209. https://doi.org/10.5194/amt-12- 169-2019

853     Grange, S. K., Carslaw, D. C., Lewis, A. C., Boleti, E., and C. Hueglin (2018). Random forest
854     meteorological normalisation models for Swiss PM 10 trend analysis, *Atmospheric Chemistry*
855     *and Physics*, 18, 6223–6239, https://doi.org/10.5194/acp-18-6223-430.

856     Grange, S. K. and D. C. Carslaw (2019). Using meteorological normalisation to detect
857     interventions in air quality time series, *Science of The Total Environment*, 653, 578–588,
858     https://doi.org/10.1016/j.scitotenv.2018.10.344

859  Grosjean, D., and J. Harrison (1985), Response of chemiluminescence NOx analyzers and
860  ultraviolet ozone analyzers to organic air pollutants, *Environ. Sci. Technol.,* 19, 862–865.

861  Hamill, T.M., J.S. Whitaker (2006), Probabilistic quantitative precipitation forecasts based on
862  reforecast analogs: theory and application, *Mon. Weather Rev.,* 134, pp. 3209-3229,
863  10.1175/MWR3237.1

864  Helfand, H. M., & Schubert, S. D. (1995). Climatology of the simulated Great Plains low-level
865  jet and its contribution to the continental moisture budget of the United States. *Journal of*
866  *Climate,* 8, 784–806.

867  Hill, C., DeLuca, C., Balaji, V., Suarez, M., & da Silva, A. (2004). Architecture of the earth
868  system modeling framework. *Computing in Science and Engineering*, 6(1), 18–28.
869  https://doi.org/10.1109/MCISE.2004.1255817

870  Honoré, C., et al. (2008), Predictability of European air quality: Assessment of 3 years of
871  operational forecasts and analyses by the PREV'AIR system, *J. Geophys. Res.*, 113, D04301,
872  doi:10.1029/2007JD008761.

873  Hu, L., Jacob, D. J., Liu, X., Zhang, Y., Zhang, L., Kim, P. S., Sulprizio, M. P., and Yantosca, R.
874  M. (2017). Global budget of tropospheric ozone: Evaluating recent model advances with satellite
875  (OMI), aircraft (IAGOS), and ozonesonde observations, *Atmos. Environ.,* 167, 323–334,
876  https://doi.org/10.1016/j.atmosenv.2017.08.036.

877  Hu, L., Keller, C. A., Long, M. S., Sherwen, T., Auer, B., Da Silva, A., Nielsen, J. E., Pawson,
878  S., Thompson, M. A., Trayanov, A. L., Travis, K. R., Grange, S. K., Evans, M. J., and Jacob, D.
879  J. (2018). Global simulation of tropospheric chemistry at 12.5 km resolution: performance and
880  evaluation of the GEOS-Chem chemical module (v10-1) within the NASA GEOS Earth system
881  model (GEOS-5 ESM), *Geosci. Model Dev.,* 11, 4603–4620, https://doi.org/10.5194/gmd-11-
882  4603-2018.

883  Ivatt, P. D. and Evans, M. J. (2020). Improving the prediction of an atmospheric chemistry
884  transport model using gradient-boosted regression trees, *Atmos. Chem. Phys.,* 20, 8063–8082,
885  https://doi.org/10.5194/acp-20-8063-2020.

886  Jacob, D. J., J. A. Logan, G. M. Gardner, R. M. Yevich, C. M. Spivakovsky, S. C. Wofsy, S.
887  Sillman, and M. J. Prather (1993), Factors regulating ozone over the United States and its export
888  to the global atmosphere, *J. Geophys. Res.,* 98, 14,817–14,826, doi:10.1029/98JD01224.

889  Jaeglé, L., Quinn, P.K., Bates, T.S., Alexander, B., Lin, J.-T. (2011). Global distribution of sea
890  salt aerosols: new constraints from in situ and remote sensing observations. *Atmospheric Chem.*
891  *Phys.* 11, 3137–3157.

892  Jaeglé, L., Shah, V., Thornton, J. A., Lopez-Hilfiker, F. D., Lee, B. H., McDuffie, E. E., Fibiger,
893  D., Brown, S. S., Veres, P., Sparks, T. L., Ebben, C. J., Wooldridge, P. J., Kenagy, H. S., Cohen,
894  R. C., Weinheimer, A. J., Campos, T. L., Montzka, D. D., Digangi, J. P., Wolfe, G. M., Hanisco,
895  T., Schroder, J. C., Campuzano-Jost, P., Day, D. A., Jimenez, J. L., Sullivan, A. P., Guo, H., and
896  Weber, R. J. (2018). Nitrogen Oxides Emissions, Chemistry, Deposition, and Export Over the
897  Northeast United States During the WINTER Aircraft Campaign, *J. Geophys. Res.-Atmos.*, 123,
898  12368-12393, https://doi.org/10.1029/2018JD029133.

899  Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G.,
900  Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P.,

901    Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M. (2015). HTAP_v2.2: a mosaic of regional
902    and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution,
903    *Atmos. Chem. Phys.,* 15, 11411–11432, https://doi.org/10.5194/acp-15-11411-2015.

904    Kang, D., R. Mathur, S.T. Rao, S. Yu (2008), Bias adjustment techniques for improving ozone
905    air quality forecasts, *J. Geophys. Res.,* 113, p. D23308, 10.1029/2008JD010151

906    Keller, C.A., Long, M.S., Yantosca, R.M., Da Silva, A.M., Pawson, S., Jacob, D.J. (2014).
907    HEMCO v1.0: a versatile, ESMF-compliant component for calculating emissions in atmospheric
908    models. *Geosci. Model Dev.* 7, 1409–1417.

909    Keller, C. A., Evans, M. J., Knowland, K. E., Hasenkopf, C. A., Modekurty, S., Lucchesi, R. A.,
910    Oda, T., Franca, B. B., Mandarino, F. C., Díaz Suárez, M. V., Ryan, R. G., Fakes, L. H., and
911    Pawson, S. (2020), Global Impact of COVID-19 Restrictions on the Surface Concentrations of
912    Nitrogen Dioxide and Ozone, *Atmos. Chem. Phys. Discuss.,* https://doi.org/10.5194/acp-2020-
913    685, in review.

914    Kleist D. T., D. F. Parrish, J. C. Derber, R. Treadon, W.-S. Wu, and S. Lord, (2009). Introduction
915    of the GSI into the NCEPs Global Data Assimilation System. *Wea. Forecasting,* 24, 1691- 1705.

916    Krol, M. and Lelieveld, J. (2003). Can the variability in tropospheric OH be deduced from
917    measurements of 1,1,1-trichloroethane (methyl chloroform)?, *J. Geophys. Res.,* 108, 4125,
918    doi:10.1029/2002JD002423.

919    Lamsal, L. N., Krotkov, N. A., Vasilkov, A., Marchenko, S., Qin, W., Yang, E.-S., Fasnacht, Z.,
920    Joiner, J., Choi, S., Haffner, D., Swartz, W. H., Fisher, B., and Bucsela, E. (2020). OMI/Aura
921    Nitrogen Dioxide Standard Product with Improved Surface and Cloud Treatments, *Atmos. Meas.
922    Tech. Discuss.*, https://doi.org/10.5194/amt-2020-200, in review.

923    Lin, S. J. (2004). A vertically Lagrangian finite-volume dynamical core for global models. *Mon.
924    Wea. Rev*., 132, 2293–2307.

925    Liu, H., Jacob, D. J., Bey, I., and Yantosca, R. M. (2001). Constraints from 210Pb and 7Be on
926    wet deposition and transport in a global three-dimensional chemical tracer model driven by
927    assimilated meteorological fields, *J. Geophys. Res.,* 106, 12109- 12128,
928    http://doi/.org/10.1029/2000jd900839.

929    Liu, F., Choi, S., Li, C., Fioletov, V.E., McLinden, C.A., Joiner, J., Krotkov, N.A., Bian, H.,
930    Janssens-Maenhout, G., Darmenov, A.S., da Silva, A.M. (2018). A new global anthropogenic
931    SO2 emission inventory for the last decade: a mosaic of satellite-derived and bottom-up
932    emissions. *Atmospheric Chem. Phys.* 18, 16571–16586. https://doi.org/10.5194/acp-18-16571-
933    2018

934    Lock, A. P., Brown, A. R., Bush, M. R., Martin, G. M., & Smith, R. N. B. (2000). A new
935    boundary layer mixing scheme. Part I: Scheme description and single-column model tests.
936    *Monthly Weather Review*, 138, 3187–3199.

937    Long, M. S., Yantosca, R., Nielsen, J. E., Keller, C. A., da Silva, A., Sulprizio, M. P., Pawson,
938    S., and Jacob, D. J. (2015). Development of a grid-independent GEOS-Chem chemical transport
939    model (v9-02) as an atmospheric chemistry module for Earth system models, *Geoscientific
940    Model Development*, 8, 595-602, http://doi/.org/10.5194/gmd-8-595-2015

941 Louis, J., & Geleyn, J. (1982). A short history of the PBL parameterization at ECMWF. In
942 Proceedings of the ECMWF workshop on planetary boundary layer parameterization (pp. 59–
943 80). Reading, UK: ECMWF.

944 Lucchesi, R. (2015). File Specification for GEOS-5 FP-IT. GMAO Office Note No. 2 (Version
945 1.4) 58 pp, available from http://gmao.gsfc.nasa.gov/pubs/office_notes.php.

946 Luo, G., Yu, F., and Schwab, J. (2019). Revised treatment of wet scavenging processes
947 dramatically improves GEOS-Chem 12.0.0 simulations of surface nitric acid, nitrate, and
948 ammonium over the United States, *Geosci. Model Dev.,* 12, 3439–3447,
949 https://doi.org/10.5194/gmd-12-3439-2019.

950 Luo, G., F. Yu, and J. Moch (2020). Further improvement of wet process treatments in GEOS-
951 Chem v12.6.0: impact on global distributions of aerosols and aerosol precursors, *Geosci. Model*
952 *Dev.,* 13, 2879-2903.

953 Mao, J., Jacob, D.J., Evans, M.J., Olson, J.R., Ren, X., Brune, W.H., Clair, J.M.St., Crounse,
954 J.D., Spencer, K.M., Beaver, M.R., Wennberg, P.O., Cubison, M.J., Jimenez, J.L., Fried, A.,
955 Weibring, P., Walega, J.G., Hall, S.R., Weinheimer, A.J., Cohen, R.C., Chen, G., Crawford, J.H.,
956 McNaughton, C., Clarke, A.D., Jaeglé, L., Fisher, J.A., Yantosca, R.M., Le Sager, P., Carouge,
957 C. (2010). Chemistry of hydrogen oxide radicals (HOx) in the Arctic troposphere in spring.
958 *Atmospheric Chem. Phys.* 10, 5823–5838. https://doi.org/10.5194/acp-10-5823-2010

959 Mao, J., Paulot, F., Jacob, D.J., Cohen, R.C., Crounse, J.D., Wennberg, P.O., Keller, C.A.,
960 Hudman, R.C., Barkley, M.P., Horowitz, L.W. (2013). Ozone and organic nitrates over the
961 eastern United States: Sensitivity to isoprene chemistry. *J. Geophys. Res.* Atmospheres 118,
962 11,256–11,268.

963 Marais, E. A., Jacob, D. J., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Krechmer,
964 J., Zhu, L., Kim, P. S., Miller, C. C., Fisher, J. A., Travis, K., Yu, K., Hanisco, T. F., Wolfe, G.
965 M., Arkinson, H. L., Pye, H. O. T., Froyd, K. D., Liao, J., and McNeill, V. F. (2016). Aqueous-
966 phase mechanism for secondary organic aerosol formation from isoprene: application to the
967 southeast United States and co-benefit of SO2 emission controls, *Atmos. Chem. Phys.,* 16, 1603-
968 1618, http://doi/.org/10.5194/acp-16-1603-2016.

969 Marais, E. and C. Wiedinmyer (2016), Air quality impact of Diffuse and Inefficient Combustion
970 Emissions in Africa (DICE-Africa), *Environ. Sci. Technol.,* 50(19), 10739–10745,
971 doi:10.1021/acs.est.6b02602.

972 Marécal, V., et al. (2015). A regional air quality forecasting system over Europe: the MACC-II
973 daily ensemble production, *Geosci. Model Dev.,* 8, 2777–2813, https://doi.org/10.5194/gmd-8-
974 2777-2015.

975 Martin, R. V., Jacob, D. J., Yantosca, R. M., Chin, M., and Ginoux, P. (2003). Global and
976 regional decreases in tropospheric oxidants from photochemical effects of aerosols, *J. Geophys.*
977 *Res.*, 108, n/a-n/a, http://doi/.org/10.1029/2002jd002622.

978 McFarlane, N. A. (1987). The effect of orographically excited gravity-wave drag on the
979 circulation of the lower stratosphere and troposphere. *Journal of the Atmospheric Sciences*, 44,
980 1775–1800.

981 McKeen S., J. Wilczak, G. Grell, I. Djalalova, S. Peckham, E.-Y. Hsie, W. Gong, V. Bouchet, S.
982 Menard, R. Moffet, J. McHenry, J. McQueen, Y. Tang, G.R. Carmichael, M. Pagowski, A. Chan,

T. Dye, G. Frost, P. Lee, R. Mathur (2005), Assessment of an ensemble of seven real-time ozone forecasts over eastern North America during the summer of 2004, *J. Geophys. Res.,* 110, p. D21307, 10.1029/2005JD005858

Miller, C., Jacob, D. J., Marais, E. A., Yu, K., Travis, K. R., Kim, P. S., Fisher, J. A., Zhu, L., Wolfe, G. M., Hanisco, T. F., Keutsch, F. N., Kaiser, J., Min, K.-E., Brown, S. S., Washenfelder, R. A., González Abad, G., and Chance, K. (2017). Glyoxal yield from isoprene oxidation and relation to formaldehyde: chemical mechanism, constraints from SENEX aircraft observations, and interpretation of OMI satellite data, *Atmos. Chem. Phys.,* 17, 8725–8738, https://doi.org/10.5194/acp-17-8725-2017.

Molod, A., Takacs, L. L., Suarez, M. J., Bacmeister, J. T., Song, I.- S., and Eichmann, A. (2012). The GEOS-5 Atmospheric General Circulation Model: Mean Climate and Development from MERRA to Fortuna. NASA Tech. Memo. 104606, Vol. 28, Tech. Rep. Series on Global Modeling and Data Assimilation, edited by: Suarez, M. J., 117 pp.

Molod, A., Partyka, G., & Suarez, M. (2013). The impact of limiting ocean roughness on GEOS-5 AGCM tropical cyclone forecasts. *Geophysical Research Letters*, 40, 411–416. https://doi.org/10.1029/2012GL053979

Molod, A., Takacs, L., Suarez, M., and Bacmeister, J. (2015). Development of the GEOS-5 atmospheric general circulation model: evolution from MERRA to MERRA2, *Geosci. Model Dev.,* 8, 1339-1356, doi:10.5194/gmd-8-1339-2015.

Molod, A., E. Hackert, Y. Vikhliaev, B. Zhao, D. Barahona, G. Vernieres, A. Borovikov, R. M. Kovach, J. Marshak, S. Schubert, Z. Li, Y.-K. Lim, L. C. Andrews, R. Cullather, R. Koster, D. Achuthavarier, J. Carton, L. Coy, J. L. M. Freire, K. M. Longo, K. Nakada, and S. Pawson, (2020). GEOS-S2S Version 2: The GMAO High Resolution Coupled Model and Assimilation System for Seasonal Prediction. *J. Geophy. Res. - Atmos.,* 125, e2019JD031767. doi: 10.1029/2019JD031767.

Monks, S. A., Arnold, S. R., Emmons, L. K., Law, K. S., Turquety, S., Duncan, B. N., Flemming, J., Huijnen, V., Tilmes, S., Langner, J., Mao, J., Long, Y., Thomas, J. L., Steenrod, S. D., Raut, J. C., Wilson, C., Chipperfield, M. P., Diskin, G. S., Weinheimer, A., Schlager, H., and Ancellet, G. (2015). Multi-model study of chemical and physical controls on transport of anthropogenic and biomass burning pollution to the Arctic, *Atmos. Chem. Phys.,* 15, 3575–3603, doi:10.5194/acp-15-3575-2015.

Montzka, S. A., Spivakovsky, C. M., Butler, J. H., Elkins, J. W., Lock, L. T., and Mondeel, D. J. (2000). New observational constraints, for atmospheric hydroxyl on global and hemispheric scales, *Science*, 288, 500–503.

Moorthi, S., & Suarez, M. J. (1992). Relaxed Arakawa Schubert: A parameterization of moist convection for general circulation models. *Monthly Weather Review*, 120, 978–1002.

Murray, L. T., D. J. Jacob, J. A. Logan, R. C. Hudman, and W. J. Koshak (2012), Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data, *J. Geophys. Res.,* 117, D20307, doi:10.1029/2012JD017934.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A.,

Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G. (2013). Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, 13, 5277–5298, doi:10.5194/acp-13-5277-2013.

Nielsen, J. E., S. Pawson, A. Molod, B. Auer, A. M. da Silva, A. R. Douglass, B. N. Duncan, Q. Liang, M. E. Manyin, L. D. Oman, W. M. Putman, S. E. Strahan, and K. Wargan (2017). Chemical Mechanisms and their Applications in the Goddard Earth Observing System (GEOS) Earth System Model. *J. Adv. Model. Earth Syst.,* 9, 3019-3044. DOI: 10.1002/2017MS001011.

Oda, T., Maksyutov, S., and Andres, R. J. (2018). The Open-source Data Inventory for Anthropogenic CO2, version 2016 (ODIAC2016): a global monthly fossil fuel CO2 gridded emissions data product for tracer transport simulations and surface flux inversions, *Earth Syst. Sci. Data*, 10, 87–107, https://doi.org/10.5194/essd-10-87-2018.

Oman, L. D., & Douglass, A. R. (2014). Improvements in total column ozone in GEOSCCM and comparisons with a new ozone-depleting substances scenario. *Journal of Geophysical Research: Atmospheres*, 119, 5613–5624. https://doi.org/10.1002/2014JD021590

Orbe, C., L. D. Oman, S. E. Strahan, D. W. Waugh, L. L. Takacs, S. Pawson, and A. M. Molod (2017). Large-Scale Atmospheric Transport in GEOS Replay Simulations. *J. Adv. Model. Earth Sy*, 9, 7, 2545-2560. DOI: 10.1002/2017MS001053.

Park, R. J., Jacob, D. J., Chin, M., and Martin, R. V. (2003). Source of carbonaceous aerosols over the United States and implications for natural visibility, *J. Geophys. Res.,* 108, 4355, https://doi.org/10.1029/2002JD003190.

Park, R. J., Jacob, D. J., Field, B. D., Yantosca, R. M., and Chin, M. (2004). Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy, *J Geophys Res-Atmos*, 109, http://doi/.org/10.1029/2003jd004473

Parrella, J.P., Jacob, D.J., Liang, Q., Zhang, Y., Mickley, L.J., Miller, B., Evans, M.J., Yang, X., Pyle, J.A., Theys, N., Van Roozendael, M. (2012). Tropospheric bromine chemistry: implications for present and pre-industrial ozone and mercury. *Atmospheric Chem. Phys.* 12, 6723–6740. https://doi.org/10.5194/acp-12-6723-2012

Patra, P., Krol, M., Montzka, S., Arnold, T., Atlas, E. L., Lintner, B., Stephens, B., Xiang, B., Elkins, J., Fraser, P., et al. (2014). Observational evidence for interhemispheric hydroxyl-radical parity, *Nature,* 513, 219.

Pawson, S., Stajner, I., Kawa, S. R., Hayashi, H., Tan, W., Nielsen, J. E., . . . Livesey, N. J. (2007). Stratospheric transport using six-hour averaged winds from a data assimilation system. *Journal of Geophysical Research*, 112, D23103. https://doi.org/10.1029/2006JD007673

Petetin, H., Bowdalo, D., Soret, A., Guevara, M., Jorba, O., Serradell, K., and Pérez García-Pando, C. (2020). Meteorology-normalized impact of the COVID-19 lockdown upon NO2 pollution in Spain, *Atmos. Chem. Phys.,* 20, 11119–11141, https://doi.org/10.5194/acp-20-11119-2020.

Pound, R. J., Sherwen, T., Helmig, D., Carpenter, L. J., and Evans, M. J. (2020). Influences of oceanic ozone deposition on tropospheric photochemistry, *Atmos. Chem. Phys.,* 20, 4227–4239, https://doi.org/10.5194/acp-20-4227-2020.

Price, C., and D. Rind (1992), A simple lightning parameterization for calculating global lightning distributions, *J. Geophys. Res.,* 97, 9919–9933, doi:10.1029/92JD00719.

Price, C., and D. Rind (1993), What determines the cloud-to-ground lightning fraction in thunderstorms, *Geophys. Res. Lett.,* 20(6), 463–466, doi:10.1029/93GL00226.

Price, C., and D. Rind (1994), Modeling global lightning distributions in a general-circulation model, *Mon. Weather Rev.,* 122(8), 1930–1939, doi:10.1175/1520-0493(1994)122<1930:MGLDIA>2.0.CO;2

Prinn, R. G., Huang, J., Weiss, R. F., Cunnold, D. M., Fraser, P. J., Simmonds, P. G., McCulloch, A., Harth, C., Salameh, P., O'Doherty, S., Wang, R. H. J., Porter, L, and Miller, B. R. (2001). Evidence for substantial variations of atmospheric hydroxyl radicals over the past two decades, *Science,* 292, 1882–1888.

Putman, W. and Lin, S.-J. (2007). Finite Volume Transport on Various Cubed Sphere Grids. *J. Comput. Phys.,* 227, 55–78. doi:10.1016/j.jcp.2007.07.022.

Pye, H.O.T., Liao, H., Wu, S., Mickley, L.J., Jacob, D.J., Henze, D.K., Seinfeld, J.H. (2009). 36 Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. *J. Geophys. Res. Atmospheres* 114, D01205. https://doi.org/10.1029/2008JD010701

Randles, C. A., and Coauthors (2017). The MERRA-2 aerosol reanalysis, 1980 onward, Part I: System description and data assimilation evaluation. *J. Climate,* 30, 6823–6850, doi:https://doi.org/10.1175/JCLI-D-16-0609.1

Ridley, D. A., Heald, C. L., and Ford, B. J. (2012). North African dust export and impacts: an integrated satellite and model perspective, *J. Geophys. Res.*, 117, D02202, https://doi.org/10.1029/2011JD016794.

Rienecker, M.M., M.J. Suarez, R. Todling, J. Bacmeister, L. Takacs, H.-C. Liu, W. Gu, M. Sienkiewicz, R.D. Koster, R. Gelaro, I. Stajner, and E. Nielsen (2008). The GEOS-5 Data Assimilation System - Documentation of Versions 5.0.1, 5.1.0, and 5.2.0. Technical Report Series on Global Modeling and Data Assimilation 104606, Vol. 27.

Rienecker and Coauthors (2011). MERRA - NASA's Modern-Era Retrospective Analysis for Research and Applications. *J. Climate,* 24, 3624-3648, doi:10.1175/JCLI-D-11-00015.1.

Rotman, D.A., Tannahill, J.R., Kinnison, D.E., Connell, P.S., Bergmann, D., Proctor, D., Rodriguez, J.M., Lin, S.J., Rood, R.B., Prather, M.J., Rasch, P.J., Considine, D.B., Ramaroson, R., Kawa, S.R. (2001). Global modeling initiative assessment model: model description, integration, and testing of the transport shell. *Journal of Geophysical Research* 106, 1669e1691.

Sandu, A., Sander, R. (2006). Technical note: Simulating chemical systems in Fortran90 and Matlab with the Kinetic PreProcessor KPP-2.1. *Atmospheric Chem. Phys.* 6, 187–195. https://doi.org/10.5194/acp-6-187-2006

Sherwen, T., Schmidt, J.A., Evans, M.J., Carpenter, L.J., Gro\s smann, K., Eastham, S.D., Jacob, D.J., Dix, B., Koenig, T.K., Sinreich, R., Ortega, I., Volkamer, R., Saiz-Lopez, A., Prados-Roman, C., Mahajan, A.S., Ordóñez, C. (2016). Global impacts of tropospheric halogens (Cl, Br, I) on oxidants and composition in GEOS-Chem. *Atmospheric Chem. Phys.* 16, 12239–12271.

Schumann, U., and H. Huntrieser (2007), The global lightning-induced nitrogen oxides source, *Atmos. Chem. Phys.,* 7(14), 3823–3907, doi:10.5194/acp-7-3823-2007.

Seinfeld, J.H. and S.N. Pandis (2016). Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. John Wiley & Sons, Hoboken.

Shah, V., Jacob, D. J., Li, K., Silvern, R. F., Zhai, S., Liu, M., Lin, J., and Zhang, Q. (2020). Effect of changing NOx lifetime on the seasonality and long-term trends of satellite-observed tropospheric NO2 columns over China, *Atmos. Chem. Phys.,* 20, 1483–1495, https://doi.org/10.5194/acp-20-1483-2020.

Shindell, D. T., et al. (2006), Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes, *J. Geophys. Res.,* 111, D19306, doi:10.1029/2006JD007100.

Sillman, S., and P. J. Samson (1995), Impact of temperature on oxidant photochemistry in urban, polluted rural and remote environments, *J. Geophys. Res.,* 100, 11,497–11,508, doi:10.1029/94JD02146.

Solomon, S., Portmann, R. W., Sasaki, T., Hofmann, D. J., and Thompson, D. W. J. (2005). Four decades of ozonesonde measurements over Antarctica, *Journal of Geophysical Research-Atmospheres*, 110, 10.1029/2005jd005917.

Spivakovsky, C. M., Logan, J. A., Montzka, S. A., Balkanski, Y. J., Foreman-Fowler, M., Jones, D. B. A., Horowitz, L. W., Fusco, A. C., Brenninkmeijer, C. A. M., Prather, M. J., Wofsy, S. C., and McElroy, M. B. (2000). Three-dimensional climatological distribution of tropospheric OH: Update and evaluation, *J. Geophys. Res.,* 105, 8931–8980.

Steinbacher, M., Zellweger, C., Schwarzenbach, B., Bugmann, S., Buchmann, B., Ordóñez, C., Prevot, A. S. H., and Hueglin, C. (2007), Nitrogen oxide measurements at rural sites in Switzerland: Bias of conventional measurement techniques, *J. Geophys. Res.,* 112, D11307, doi:10.1029/2006JD007971.

Streets, D.G., Canty T., Carmichael, G.R. , de Foy, B. Dickerson, R.R, Duncan, B.N., Edwards, D.P. , Haynes, J.A., Henze, D.K., Houyoux, M.R., Jacob, D.J., Krotkov, N.A., Lamsal, L.N., Liu, Y., Lu, Z., Martin, R.V., Pfister, G.G., Pinder, R.W., Salawitch, R.J., and K.J. Wecht (2013), Emissions estimation from satellite retrievals: A review of current capability, *Atmospheric Environment,* Volume 77, Pages 1011-1042, https://doi.org/10.1016/j.atmosenv.2013.05.051.

Strode, S. A., Duncan, B. N., Yegorova, E. A., Kouatchou, J., Ziemke, J. R., and Douglass, A. R. (2015). Implications of carbon monoxide bias for methane lifetime and atmospheric composition in chemistry climate models, *Atmos. Chem. Phys.,* 15, 11789–11805, https://doi.org/10.5194/acp-15-11789-2015.

Stettler, M., Eastham, S., and Barrett, S, (2011). Air quality and public health impacts of UK airports. Part I: Emissions, *Atmos. Environ.,* 45, 5415–5424, doi:10.1016/j.atmosenv.2011.07.012, 2011.

Strahan, S.E., B.N. Duncan and P. Hoor (2007). Observationally-derived diagnostics of transport in the lowermost stratosphere and their application to the GMI chemistry transport model , *Atmos. Chem. Phys.,* 7, 2435-2445.

Suarez, M., Trayanov, A., Hill, C., Schopf, P., & Vikhliaev, Y. (2007). MAPL: A high-level programming paradigm to support more rapid and robust encoding of hierarchical trees of interacting high-performance components. In Proceedings of the 2007 symposium on component and framework technology in high-performance and scientific computing (pp. 11–20). https://doi.org/10.1145/1297385.1297388

Tarasova, O. A., and A. Y. Karpetchko (2003), Accounting for local meteorological effects in the ozone time-series of Lovozero (Kola Peninsula), *Atmos. Chem. Phys.,* 3(4), 941–949, doi:10.5194/acp-3-941-2003.

Thompson, A. M., Witte, J. C., Sterling, C., Jordan, A., Johnson, B. J., Oltmans, S. J., Fujiwara, M., Vomel, H., Allaart, M., Piters, A., Coetzee, G. J. R., Posny, F., Corrales, E., Diaz, J. A., Felix, C., Komala, N., Lai, N., Nguyen, H. T. A., Maata, M., Mani, F., Zainal, Z., Ogino, S. Y., Paredes, F., Penha, T. L. B., da Silva, F. R., Sallons-Mitro, S., Selkirk, H. B., Schmidlin, F. J., Stubi, R., and Thiongo, K. (2017). First Reprocessing of Southern Hemisphere Additional Ozonesondes (SHADOZ) Ozone Profiles (1998-2016): 2. Comparisons With Satellites and Ground-Based Instruments, *Journal of Geophysical Research-Atmospheres,* 122, 13000-13025, 10.1002/2017jd027406.

Travis, K. R., Jacob, D. J., Fisher, J. A., Kim, P. S., Marais, E. A., Zhu, L., Yu, K., Miller, C. C., Yantosca, R. M., Sulprizio, M. P., Thompson, A. M., Wennberg, P. O., Crounse, J. D., St. Clair, J. M., Cohen, R. C., Laughner, J. L., Dibb, J. E., Hall, S. R., Ullmann, K., Wolfe, G. M., Pollack, I. B., Peischl, J., Neuman, J.A., and Zhou, X. (2016). Why do models overestimate surface ozone in the Southeast United States?, *Atmos. Chem. Phys.,* 16, 13561–13577, https://doi.org/10.5194/acp-16-13561-2016.

Travis, K. R. and Jacob, D. J. (2019). Systematic bias in evaluating chemical transport models with maximum daily 8 h average (MDA8) surface ozone for air quality applications: a case study with GEOS-Chem v9.02, *Geosci. Model Dev.,* 12, 3641–3648, https://doi.org/10.5194/gmd-12-3641-2019.

Wang, Q., Jacob, D.J., Spackman, J.R., Perring, A.E., Schwarz, J.P., Moteki, N., Marais, E.A., Ge, C., Wang, J., Barrett, S.R.H. (2014). Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific. *J. Geophys. Res. Atmospheres* 119, 195–206.

Wang, X., D.J. Jacob, M.P. Sulprizio, S.D. Eastham, L. Zhu, Q. Chen, B. Alexander, T. Sherwen, M.J. Evans, B.H. Lee, J.D. Haskins, F.D. Lopez-Hilfike, J.A. Thornton, G.L. Huey, and H. Liao (2019), The role of chlorine in global tropospheric chemistry, *Atmos. Chem. Phys.*, 19, 3981-4003.

Wargan, K., S. Pawson, M. A. Olsen, J. C. Witte, A. R. Douglass, J. R. Ziemke, S. E. Strahan, and J. E. Nielsen (2015). The Global Structure of Upper Troposphere-Lower Stratosphere Ozone in GEOS-5: A Multi-Year Assimilation of EOS Aura Data. *J. Geophys. Res. - Atmos*, 120, 2013-2036. doi: 10.1002/2014JD022493.

Wargan, K., N. Kramarova, B. Weir, S. Pawson, and S. Davis (2020). Towards a reanalysis of stratospheric ozone for trend studies: Assimilation of the Aura Microwave Limb Sounder and Ozone Mapping and Profiler Suite Limb Profiler data. *J. Geophys. Res. - Atmos.,* 125, e2019JD031892. doi: 10.1029/2019JD031892.

1189    Waugh, D. W., & Eyring, V. (2008). Quantitative performance metrics for stratospheric-
1190    resolving chemistry-climate models. *Atmospheric Chemistry and Physics*, 8, 5699–5713.
1191    https://doi.org/10.5194/acp-8-5699-2008

1192    Wesely, M. L. (1989). Parameterization of Surface Resistances to Gaseous Dry Deposition in
1193    Regional-Scale Numerical-Models, *Atmos. Environ.,* 23, 1293-1304, http://doi/.org/Doi
1194    10.1016/0004-6981(89)90153-4.

1195    Wilczak, J., S.A. McKeen, I. Djalalova, et al. (2006). Bias-corrected ensemble and probabilistic
1196    forecasts of surface ozone over eastern North America during the summer of 2004, *J. Geophys.*
1197    *Res.*, 111 (D23S28), 10.1029/2006JD007598

1198    Winer, A. M., J. W. Peters, J. P. Smith, and J. N. Pitts Jr. (1974), Response of commercial
1199    chemiluminescence NO–NO2 analyzers to other nitrogen-containing compounds, *Environ. Sci.*
1200    *Technol.,* 8, 1118–1121.

1201    Wu, W.-S., R.J. Purser and D.F. Parrish (2002). Three-dimensional variational analysis with
1202    spatially inhomogeneous covariances. *Mon. Wea. Rev.,* 130, 2905-2916.

1203    Zhang, Y., Jacob, D. J., Lu, X., Maasakkers, J. D., Scarpelli, T. R., Sheng, J.-X., Shen, L., Qu,
1204    Z., Sulprizio, M. P., Chang, J., Bloom, A. A., Ma, S., Worden, J., Parker, R. J., and Boesch, H.
1205    (2020), Attribution of the accelerating increase in atmospheric methane during 2010–2018 by
1206    inverse analysis of GOSAT observations, *Atmos. Chem. Phys. Discuss.*,
1207    https://doi.org/10.5194/acp-2020-964, in review.