

Development of the CCpp-Based GEFS-Aerosols Component in the Unified Forecast System for Subseasonal Prediction (UFS-Chem v1.0)

Li Zhang^{1,2}, Haiqin Li^{1,2}, Georg A. Grell*, Partha S. Bhattacharjee³, Gonzalo A. Ferrada^{1,2}, Benjamin W. Green^{1,2}, Shan Sun², Ligia Bernardet², Anders Jensen², Barry Baker⁴, Li Pan³, Jian He^{1,5}, Jordan Schnell^{1,2}, Ravan Ahmadov², Samuel Trahan^{1,2}, Dustin Swales², Anning Cheng³, Fanglin Yang⁴, Rebecca H. Schwantes⁵, Brian C. McDonald⁵, Dominikus Heinzeller^{6,7}, Shobha Kondragunta⁸

¹CIRES, University of Colorado Boulder, Boulder, CO, US;

²Global Systems Laboratory, NOAA, Boulder, CO, US;

³SAIC/Lynker at NOAA/NWS/OMD, College Park, MD, US;

⁴NOAA/NWS/OMD, College Park, MD, US;

⁵Chemical Sciences Laboratory, NOAA, Boulder, CO, US;

⁶Marine Meteorology Division, Naval Research Laboratory, Monterey, CA, US

⁷Cooperative Programs for the Advancement of Earth System Sciences, University Corporation for Atmospheric Research, Boulder, CO, US

⁸NOAA/NESDIS Center for Satellite Applications and Research, College Park, MD, US;

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Correspondence to: Li Zhang (kate.zhang@noaa.gov)
CIRES, University of Colorado Boulder & GSL, NOAA
325 Broadway David Skaggs Research Center R/GSL1
Boulder, CO 80305
1-307-240-9036

^{1*} Retired from NOAA Global Systems Laboratory, currently a visitor at the National Science Foundation National Center for Atmospheric Research (NCAR)

33 **Abstract.**

34 The Global Ensemble Forecast System (GEFS) version 12 has been operational at the National Centers for Environmental
35 Prediction since September 2020, with GEFS-Aerosols serving as its global aerosol forecasting member. In 2023, GEFS-Aerosols
36 was upgraded to version 12.3 in operations, incorporating improvements to wet deposition, anthropogenic and biomass burning
37 emissions, aerosol optical depth calculations, and the FENGSHA dust emission scheme. While GEFS-Aerosols provides valuable
38 operational aerosol forecasts on medium-range timescales, its one-way coupling strategy restricts aerosol–atmosphere interactions
39 to prescribed climatological fields, thereby precluding fully interactive aerosol feedbacks for extended-range prediction. To
40 overcome this limitation, we develop the Unified Forecast System coupled with Chemistry (UFS-Chem), which incorporates the
41 Configurable Atmospheric Chemistry (CATChem) library and modeling component to include both aerosol and gas-phase
42 chemistry schemes. In the current UFS-Chem configuration, the aerosol component is based on the operational GEFS-Aerosols
43 v12.3 and implemented using the Common Community Physics Package (CCPP) framework. Relative to GEFS-Aerosols v12.3,
44 UFS-Chem incorporates several enhancements, including inline aerosol radiative feedback, inline large-scale wet deposition, and
45 updated FENGSHA dust scheme. In particular, the integration of inline large-scale wet deposition and aerosol indirect effects
46 through the Thompson aerosol-aware microphysics scheme improves the representation of aerosol–cloud interactions and supports
47 weather and subseasonal-to-seasonal forecasting when fully coupled with ocean, sea ice, land, and wave components. The
48 performance of UFS-Chem in weather and subseasonal prediction is evaluated against reanalysis data, ground-based
49 measurements, and satellite observations. The relative impacts of aerosol radiative and indirect effects on weather and subseasonal
50 predictions are investigated and quantified, advancing our understanding of how aerosol–radiation and aerosol–cloud interactions
51 influence forecast skill across timescales.

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1. Introduction

Aerosols and reactive gases play a fundamental role in the Earth system through their effects on radiation, clouds, precipitation, and atmospheric composition, with consequences for both climate and human health (Boucher et al., 2013; IPCC, 2021). Accurate forecasts of atmospheric composition on weather-to-seasonal timescales are essential for anticipating air quality events, dust outbreaks, and long-range transport of pollution and biomass burning plumes, with direct applications to aviation safety, renewable energy, and public health (Grell and Baklanov, 2011; Zhang et al., 2014). Over the past two decades, substantial progress has been made in operational aerosol and chemistry forecasting, with international systems demonstrating the benefits of coupling atmospheric composition with numerical weather prediction. Notable examples include the National Aeronautics and Space Administration (NASA)’s Goddard Chemistry Aerosol Radiation and Transport (GOCART) model (Chin et al., 2000), which provided one of the first global representations of aerosol processes, and Goddard Earth Observing System Forward Processing (GEOS-FP), a global meteorological analysis and forecasting system that provides the weather fields supporting aerosol and atmospheric composition modeling (Rienecker et al., 2008; Molod et al., 2015). Building upon the GEOS-FP framework, the Goddard Earth Observing System Composition Forecast (GEOS-CF) system was subsequently developed to integrate comprehensive atmospheric chemistry and delivers global operational forecasts of aerosols and atmospheric composition (Colarco et al., 2010; Keller et al., 2021). In Europe, the Copernicus Atmosphere Monitoring Service (CAMS) integrates aerosol and chemistry modules into the ECMWF Integrated Forecasting System, assimilating satellite data to provide skillful composition forecasts (Flemming et al., 2015; Inness et al., 2019). Studies at the German Weather Service (DWD) using the Consortium for Small-scale Modeling – Aerosols and Reactive Trace gases (COSMO-ART) and the ICOSahedral Nonhydrostatic model – Aerosols and Reactive Trace gases (ICON-ART) have demonstrated that accounting for dust direct and indirect effects can improve surface radiation and weather forecasts during Saharan dust outbreaks (Bangert et al., 2012; Rieger et al., 2017). In the United States, the Rapid Refresh Forecast System – Smoke and Dust (RRFS-SD) demonstrated that including smoke aerosol direct feedback improves near-surface temperature, wind speed, and visibility forecasts (Li et al., 2025). These systems underscore the importance of inline aerosol–radiation and aerosol–cloud interactions, as well as data assimilation, for predictive skill.

In the United States, the National Centers for Environmental Prediction (NCEP) implemented GEFS-Aerosols as a member of GEFS v12 operations in September 2020 (Zhang et al., 2022; Bhattacharjee et al., 2023; Li et al., 2024). GEFS-Aerosols represents the second operational global aerosol forecasting capability within NOAA, replacing the NEMS Global Forecast System Aerosol Component version 2 (NGACv2), and supports applications including dust forecasting, wildfire aerosol transport, and the provision of boundary conditions for regional models. With the transition to GEFS-Aerosols v12.3 in 2023, major updates were introduced when compared to GEFS-Aerosols v12, including improved wet deposition processes, anthropogenic and biomass burning emissions, aerosol optical depth (AOD) calculations, and an updated FENGSHA dust emission scheme.

The aerosol component in GEFS-Aerosols is based on the GOCART model, a simplified aerosol scheme that does not include full gas-phase or stratospheric chemistry. To overcome this limitation, we have initiated the development of the Configurable ATmospheric Chemistry (CATChem) library and modeling component that includes comprehensive aerosol and chemistry processes to represent atmospheric composition within the Unified Forecast System (UFS; Jacobs, 2021) framework, named as UFS-Chem. A detailed description of the CATChem framework is provided in He et al. (2026). In CATChem v1, the aerosol component from GEFS-Aerosols v12.3 (GEFS-GOCART) and the gas-phase chemistry from Atmosphere Model version 4.1 (AM4.1) developed by the Geophysical Fluid Dynamics Laboratory (GFDL) have been implemented as configurable options.

In the initial GEFS-Aerosols configuration, atmospheric composition processes were coupled to the dynamical core and physics suite through the National Unified Operational Prediction Capability (NUOPC) framework, in which chemistry and aerosol modules were treated largely as external components. While this approach provided modular flexibility, it resulted in relatively

108 loose coupling between chemistry and physics. Many key aerosol processes — such as emissions, transport, wet and dry deposition,
109 microphysics, and radiation — were executed within the chemistry or aerosol component using parameterizations independent of
110 those in the host atmospheric physics. Most chemistry and aerosol models include their own representations of convective
111 transport, convective wet deposition, planetary boundary layer (PBL) mixing, dry deposition, and large-scale wet removal (e.g.,
112 GOCART, Chin et al., 2000; CMAQ, Byun and Schere, 2006; GEOS-Chem, Bey et al., 2001). These parameterizations often differ
113 in numerical formulation, underlying assumptions, and process behavior from the corresponding physics schemes in the host
114 atmospheric model. Consequently, chemical and aerosol tracers may experience transport, scavenging, and removal processes that
115 are inconsistent with those applied to meteorological variables, leading to discrepancies in process coupling, temporal splitting,
116 and physical interpretation. However, atmospheric chemistry and aerosols are inherently interactive and strongly coupled with
117 physical processes. Aerosol emissions depend on land surface conditions and PBL turbulence; chemical tracers are transported by
118 PBL mixing and convective processes; wet scavenging is controlled by cloud microphysics and precipitation; and aerosol–radiation
119 interactions directly modify atmospheric thermodynamics and circulation. Treating these processes separately within standalone
120 chemistry or aerosol components can therefore limit the fidelity of chemistry–physics feedbacks.

121 To achieve a more physically consistent representation of these interactions, we initiated in 2021 an alternative coupling approach
122 for both global and regional modeling applications, in which the aerosol and chemistry components are decomposed into process-
123 level modules and embedded directly within the physics suite of the host model through the Common Community Physics Package
124 (CCPP) infrastructure (Heinzeller et al., 2023, Bernardet et al., 2024). This coupling approach was first applied in an intermediate
125 configuration, integrating the aerosol emission and GEFS-GOCART aerosol modules from GEFS-Aerosols v12 for weather, and
126 subseasonal to seasonal (S2S) prediction (Li et al., 2024; Sun et al., 2025). The present study builds upon those efforts with a more
127 comprehensive and optimized implementation, coupling the updated GEFS-GOCART module from GEFS-Aerosols v12.3 with
128 GFSv17 physics in UFS-Chem, which incorporates additional enhancements including inline large-scale wet deposition and
129 aerosol indirect effects. Meanwhile, the CCPP coupling infrastructure was used beyond aerosol-only modeling to support the full
130 gas-phase chemistry capability of GFDL's AM4.1, as described in He et al. (2026). At the regional scale, the same approach has
131 been adopted in the development of the RRFS-SD v1, in which smoke plume rise and dust emission modules are embedded as
132 CCPP physics subroutines within the RRFS for convection-allowing smoke and dust prediction over North America (Li et al.,
133 2025). By integrating aerosol and gas-phase chemistry processes as CCPP-compliant physics subroutines, this approach enables
134 emissions, transport, scavenging, and radiative interactions to share the same numerical schemes, time stepping, and state variables
135 as the host model physics, thereby providing tighter and more physically consistent coupling between atmospheric composition
136 and model physics.

137 While the updates in GEFS-GOCART of GEFS-Aerosols v12.3 enhanced forecast skill, the system remained constrained by its
138 coupling strategy, which permitted only aerosol–radiation feedback of direct and semi-direct effects through prescribed
139 climatological aerosol fields. Interactive aerosol–atmosphere feedbacks — known to be critical for realistic simulations of weather
140 and S2S variability (Benedetti et al., 2018; Freire et al., 2020) — were not represented. To address this limitation, a two-way
141 coupling strategy is required in which the inline-predicted aerosol fields interact with meteorological processes through both
142 radiation and cloud microphysics. Recent studies have made progressive steps toward incorporating aerosol feedback within the
143 UFS framework. Cheng and Yang (2023) demonstrated the impact of aerosol direct radiative effects on numerical weather forecasts
144 by replacing the legacy Optical Properties of Aerosols and Clouds aerosol climatology with second Modern-Era Retrospective
145 Analysis for Research and Applications (MERRA-2) climatological aerosol fields in the GFSv16 physics suite, showing
146 improvements in radiative flux biases, circulation, and precipitation over monsoon regions. Sun et al. (2025) investigated the
147 aerosol radiative feedbacks of direct and semi-direct effects on subseasonal prediction using the UFS with GFSv16 physics, fully

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coupled with ocean, sea ice, land, and wave components, together with an earlier version of the GEFS-GOCART module based on the operational GEFS-Aerosols v12. Cheng et al. (2026) evaluated aerosol–cloud–radiation interactions by coupling both MERRA-2 climatological and MERRA-2 analysis aerosols with radiation and the Thompson microphysics scheme, enabling the representation of aerosol indirect effects in addition to the direct and semi-direct effects in medium-range weather predictions during a fire event. Their results revealed that the inclusion of aerosol–cloud interactions produces additional net cooling at the top of the atmosphere (TOA) and modifies the vertical distribution of cloud liquid water and ice water content. While these studies have established the individual benefits of aerosol radiative ~~feedback and aerosol indirect effects~~, a comprehensive assessment of their combined and relative impacts on subseasonal prediction within a fully coupled UFS-Chem framework — using inline-predicted rather than climatological aerosol fields or diagnostic aerosol from MERRA-2 reanalysis data for both radiative and microphysical interactions — has not yet been undertaken. The present study addresses this gap by systematically quantifying the contributions of aerosol radiative feedback and ~~the interactive aerosol–cloud–meteorology coupling associated with aerosol indirect effects~~, using the improved GEFS-GOCART schemes, both individually and in combination, to weather and subseasonal forecast skill.

The design of UFS-Chem offers several distinct advantages for advancing aerosol-aware prediction across timescales. The CCPP-based coupling infrastructure supports a range of atmospheric composition mechanisms, from the simplified GOCART aerosol scheme to the comprehensive AM4.1 gas-phase chemistry, within a unified framework through the CATChem library and modeling component, providing the flexibility to configure the level of chemical complexity appropriate for different applications. As a modular library, CATChem can also be readily adapted to other host models beyond the UFS, broadening its applicability across the modeling community. This modular design ensures consistency of aerosol–physics coupling across UFS weather, subseasonal, and seasonal applications, avoiding the need for separate development paths at different timescales. Building upon this infrastructure, UFS-Chem enables interactive aerosol direct, semi-direct, and indirect effects through two-way coupling between the inline-predicted aerosol fields and the host model radiation and microphysics schemes.

In the present study, we describe the development of UFS-Chem with the aerosol component from GEFS-Aerosols v12.3 for weather and subseasonal prediction. Section 2 describes the observational, reanalysis, and model data used for evaluation, along with the UFS and GEFS-GOCART configurations and their integration within the CCPP infrastructure. Section 3 shows the improved performance of GEFS-Aerosols v12.3. The implementation of inline large-scale wet deposition into the Thompson microphysics scheme is presented in Section 4. Section 5 presents the ~~implementation of aerosol indirect effects through the interactive coupling between GOCART aerosols and the Thompson microphysics scheme~~. The aerosol radiative ~~feedback and indirect effects~~ in subseasonal prediction are quantified in Section 6. A summary, discussion, and future plans are provided in Section 7.

2. Data and model

2.1 Observational, reanalysis, and ensemble analysis data

A variety of observational datasets, reanalysis and ensemble analysis products are used in this study to evaluate the performance of the model prediction and to provide reference fields for comparison. These datasets are as follows:

- 1) Total AOD fields from MERRA-2 (Gelaro et al., 2017) are used as the primary atmospheric reanalysis reference in this study. MERRA-2 includes extensive assimilation of aerosol observations and provides time-averaged (1-h) AOD reanalysis products at $0.625^\circ \times 0.5^\circ$ horizontal resolution on 72 vertical levels.
- 2) Satellite-based AOD observations from the Moderate Resolution Imaging Spectroradiometer (MODIS) are employed for independent evaluation. We use the MODIS Collection 6.1 Level-3 daily and monthly AOD dataset at 1° horizontal resolution

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from the Aqua satellite (Levy et al., 2013), which is available from NASA Level-1 and Atmosphere Archive and Distribution System (LAADS) Distributed Active Archive Center (DAAC, <https://ladsweb.nascom.nasa.gov/>). Collection 6.1 incorporates refined retrieval algorithms, including the expanded Deep Blue algorithm (Hsu et al., 2013; Sayer et al., 2013), and introduces a merged AOD product that combines retrievals from the Dark Target and Deep Blue algorithms to produce a spatially consistent dataset covering surface types ranging from oceans to bright deserts (Sayer et al., 2014). In this work, the Dark_Target_Deep_Blue_Combined_Mean product at 550 nm is used for quantitative evaluation of model results. The expected errors for Dark Target retrievals at 10 km resolution are $\pm (0.05 + 15 \% \text{ AOD})$ over land and $\pm (0.03 + 5 \% \text{ AOD})$ over ocean. For the Deep Blue retrievals at 10 km resolution over land, the expected errors are approximately $\pm (0.03 + 21 \% \text{ AOD})$ for arid surfaces and $\pm (0.03 + 18 \% \text{ AOD})$ for vegetated surfaces (Levy et al., 2013).

- 3) Additional satellite AOD observations are obtained from the Visible Infrared Imaging Radiometer Suite (VIIRS) sensor onboard the Suomi National Polar-orbiting Partnership satellite, which provides aerosol environmental data records based on daily global observations (Jackson et al., 2013; Liu et al., 2013). Beginning in 2012, VIIRS provides AOD at 550 nm on a global 0.1° horizontal resolution grid, and the daily gridded data are available from NOAA/NESDIS (https://www.star.nesdis.noaa.gov/atmospheric-composition-training/satellite_data.php#viirs_star).
- 4) Ground-based AOD measurements from the Aerosol Robotic Network (AERONET; Holben et al., 1998) level 1.5 are used for point-based validation. AERONET is a globally distributed network of automated sun-photometer measurements comprising over 700 stations, providing AOD, surface solar flux, and additional radiometric products. The network employs CIMEL sun-sky spectral radiometers, which measure direct sun radiances at eight spectral channels centered at 340, 380, 440, 500, 675, 870, 940, and 1020 nm. AOD uncertainties in the direct sun measurements are within ± 0.01 for wavelengths longer than 440 nm and ± 0.02 for shorter wavelengths (Eck et al., 1999).
- 5) Global satellite-derived estimates of TOA net radiative flux and cloud fraction were obtained from Edition 4.2 of the Clouds and the Earth's Radiant Energy System (CERES) Energy Balanced and Filled (EBAF) products (Loeb et al., 2018; Kato et al., 2018; NASA/LARC/SD/ASDC, 2023). CERES EBAF data (hereafter simply "CERES data") are available as monthly means on a $1^\circ \times 1^\circ$ grid.
- 6) Version 2.1 of the Daily Optimum Interpolation Sea Surface Temperature (OISST; Huang et al., 2021) combines buoy and ship data with satellite measurements to provide a daily record of sea-surface temperature (SST) at 0.25° horizontal resolution. For the purposes of this study, OISST data was converted to monthly averages and regridded to 1° horizontal resolution.
- 7) The International Cooperative for Aerosol Prediction Multi-Model Ensemble (ICAP-MME; Reid et al., 2011; Sessions et al., 2015; Xian et al., 2019) provides 6-hourly global forecasts of total and dust AOD out to 120 h at 1° horizontal resolution. The total AOD ensemble is derived from four core multi-species models of ECMWF-CAMS, Japan Meteorological Agency Model of Aerosol species in the Global Atmosphere (JMA-MASINGAR), NASA Goddard Earth Observing System Version 5 (GEOS-5) and the Naval Research Lab Navy Aerosol Analysis and Prediction System (NRL-NAAPS), all of which incorporate aerosol data assimilation and satellite-based smoke emissions. Dust-only AOD additionally includes contributions from Barcelona Supercomputer Center Chemical Transport Model (NMMB/BSC-CTM), United Kingdom Met Office Unified Model (UKMO-UM), and NGACv2, although NGACv2 is excluded from the total AOD ensemble mean.

2.2 Key updates in operational GEFS-Aerosols v12.3

Table 1 shows the relationship and comparison of the global aerosol forecast systems: GEFS-Aerosols v12, GEFS-Aerosols v12.3, and UFS-Chem (this study). In the operational GEFS-Aerosols v12 and v12.3 configurations, the aerosol module employs a one-way coupling with the host model (FV3 with GFSv15 physics), whereby the prognostic aerosol fields from the coupled aerosol

model do not feed back into the radiation calculations. Instead, MERRA-2 climatological aerosol data are used to compute aerosol radiative effects, including direct and semi-direct effects (Cheng and Yang, 2023). This configuration does not allow aerosol radiative feedback associated with real-time aerosol variability. It is worth noting that even within the NUOPC framework, GEFS-Aerosols v12 and v12.3 already achieves physically consistent aerosol convective transport, wet deposition, and boundary-layer mixing by leveraging the Simplified Arakawa-Schubert (SAS) convection scheme and PBL parameterization within the GFS v15 physics suite (Zhang et al., 2022). While convective transport and PBL mixing benefit from consistent physics in GEFS-Aerosols v12.3, other processes such as aerosol–radiation interactions, dry deposition, emissions coupling, and microphysics-dependent removal are still handled outside the physics suite.

Several key updates were introduced in GEFS-Aerosols v12.3, including (1) an update of anthropogenic emissions from the Community Emissions Data System (CEDS) 2014 inventory to the 2019 version, (2) revised parameterizations for large-scale wet deposition and aerosol scavenging, and (3) fixes to known issues in the AOD calculation and the FENGSHA dust emission scheme. GEFS-Aerosols v12.3 has been implemented operationally since early 2023. Together, these updates aim to improve the representation of aerosol loading and reduce systematic biases in short-range forecasts. These updates have also been incorporated into UFS-Chem.

2.3 CCPP-Based GEFS-Aerosols v12.3

As shown in Table 1, UFS-Chem couples the aerosol model from GEFS-Aerosols v12.3, but it is implemented using the CCPP infrastructure with UFS. Figure 1 illustrates the differences in coupling structure between the NUOPC-based GEFS-Aerosols v12.3 configuration and the CCPP-based implementation adopted in UFS-Chem. In the NUOPC framework, atmospheric physics and chemistry are organized as separate components. Physical parameterizations—including radiation, land surface processes, PBL mixing, convection, and microphysics—are executed within the atmospheric physics component, while emissions, aerosol processes, chemistry, and deposition are handled within a chemistry or aerosol driver. Information exchange between these components occurs through predefined interfaces, resulting in a largely sequential execution of physics and chemistry processes. In contrast, the CCPP-based implementation reorganizes the coupling strategy by embedding chemistry and aerosol processes directly within the physics time step. As shown in Fig. 1, emissions processes of anthropogenic, biogenic, volcanic, dust, sea salt, fire, and dimethyl sulfide sources (DMS), aerosol gravity settling, dry deposition, large-scale wet deposition, and aerosol optical property calculations are treated as physics-aware modules that interact directly with radiation, PBL mixing, convection, and microphysics. Convective scavenging and convective transport continue to be handled by SAS convection scheme. This design allows chemical and aerosol tracers to be transported, mixed, scavenged, and removed using the same physical parameterizations applied to meteorological tracers.

In the CCPP framework, chemistry and aerosol processes are decomposed into process-level modules and embedded directly within the GFS physics suite, allowing tracer transport, mixing, scavenging, deposition, and aerosol–radiation interactions to be handled within the host physics time step. Under this framework, tracer transport by PBL mixing and convection, convective and stratiform wet scavenging, and microphysics-dependent removal are calculated within the host physics parameterizations rather than being recomputed inside a standalone chemistry or aerosol model. This approach allows aerosols and gases to experience atmospheric processes using the same physical tendencies applied to meteorological fields. In addition, because the meteorological fields required for transport and deposition are already available within the physics suite, there is no need to import or re-diagnose these fields within the chemistry or aerosol component. As a result, redundant calculations may be reduced, and chemistry–physics interactions can be handled inline within the physics workflow. Using the same physics suite and model configuration as GEFS-Aerosols v12.3, the CCPP-based GEFS-Aerosols implementation at the initial stage of development required approximately 22–

276 23 min for a 1-day forecast, compared with approximately 24 min for GEFS-Aerosols v12.3, corresponding to a 4–8% reduction
277 in runtime.

278 Overall, the CCpp-based GEFS-Aerosols configuration in UFS-Chem provides a tightly coupled and extensible framework for
279 atmospheric composition modeling. This design facilitates the representation of chemistry–physics interactions, supports future
280 expansion to alternative chemical mechanisms, and aligns GEFS-Aerosols with the unified physics architecture of the broader
281 UFS.

282 2.4 UFS-Chem configuration

283 The UFS is a fully coupled modeling framework capable of integrating multiple Earth system components for subseasonal
284 prediction (Fu et al., 2024). In this study, the UFS configuration includes the atmospheric model employing the Finite-Volume
285 Cubed-Sphere (FV3) dynamical core (Harris et al., 2021), the GFS v17 physics suite, the ocean model (GFDL Modular Ocean
286 Model version 6, MOM6; Adcroft et al., 2019), the sea-ice model (Los Alamos Sea Ice Model version 6, CICE6), the wave model
287 (WAVEWATCH III, WW3; WAVEWATCH III Development Group, 2019), and CATChem v1 (He et al., 2026). The atmospheric
288 model is configured with a horizontal grid of C384 (approximately 25 km horizontal resolution) and employs 127 vertical levels,
289 with the model top at 0.01 hPa, providing enhanced vertical resolution in the upper atmosphere. The MOM6 is configured at 25-
290 km horizontal resolution with 75 vertical layers, while CICE6 also operates at a 25-km horizontal resolution. WW3 uses a 50-km
291 horizontal resolution. Key updates in the GFS v17 physics suite include the Thompson microphysics scheme (Thompson et al.,
292 2008; Thompson and Eidhammer, 2014), the Noah-MP land surface model (Niu et al., 2011, He et al., 2023), the addition of non-
293 orographic gravity wave drag, and updates to the scale-aware SAS convection scheme (Han and Pan, 2011, Bengtsson and Han,
294 2024) and the turbulent kinetic energy–based eddy-diffusivity mass-flux PBL scheme (Han et al., 2022).

295 For the aerosol model configuration in UFS-Chem, the biomass-burning emissions are prescribed using the NOAA operational
296 Blended Global Biomass Burning Emissions Product version 4 dataset (GBBEPx v4), which has been used for real-time operational
297 applications since 2019. The GBBEPx v4 system provides daily global emissions of particulate matter with aerodynamic diameter
298 $\leq 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$), black carbon (BC), organic carbon (OC), carbon monoxide (CO), carbon dioxide (CO_2), and sulfur dioxide (SO_2)
299 (Zhang et al., 2019). A one-dimensional, time-dependent plume-rise module from the High-Resolution Rapid Refresh Smoke
300 (HRRR-Smoke) model has been implemented in UFS-Chem, similar to that in GEFS-Aerosols, to calculate biomass burning
301 emission injection heights and emission rates online (Ahmadov et al., 2017). The scheme is a modified version of the WRF-Chem
302 plume-rise module (Freitas et al., 2007) that uses fire radiative power to estimate fire heat fluxes instead of a lookup table. It is
303 applied with the GBBEPx v4 biomass burning emissions dataset to represent plume-rise processes that vertically distribute biomass
304 burning emissions. Global anthropogenic emissions are taken from the CEDS v2019, which supplies emissions of BC, OC, and
305 SO_2 (McDuffie et al., 2020) for GEFS-GOCART aerosols. Compared with CEDS v2014 (Hoesly et al., 2018), this updated dataset
306 extends emissions through 2017 and improves consistency with other widely used global bottom-up inventories. The GEFS-
307 GOCART aerosol mechanism is originally from WRF-Chem GOCART (Zhang et al., 2022), which employs a bulk representation
308 for sulfate, OC, and BC, distinguishing between hydrophilic and hydrophobic components. Dust aerosols are represented using the
309 sectional FENGSHA dust scheme developed by NOAA’s Air Resources Laboratory (Zhang et al., 2022). Relative to the version
310 implemented in GEFS-Aerosols v12.3, the dust emission parameterization has been updated by incorporating the Baker–
311 Schepanski map (NOAA EPIC, 2023) and improved surface input datasets. The sea salt scheme uses a five-bin sectional approach
312 based on Gong (2003), with modifications similar to those of Jaeglé et al. (2011). Sea salt emissions are parameterized as a function
313 of near-surface wind speed and do not directly depend on wave model coupling. Similar to GEFS-Aerosols v12.3, aerosol
314 convective transport, wet deposition, and boundary-layer mixing are handled consistently through the SAS convection scheme and

PBL parameterization within the GFS v17 physics suite. In this study, the UFS-Chem experiments are configured using the High-Resolution 3 configuration, with details provided in He et al. (2026). In the current UFS-Chem framework, a two-way inline coupling has been implemented between the GOCART aerosol model and the radiation scheme, enabling aerosol radiative feedback of direct and semi-direct effects from the inline-coupled aerosol simulation. The AOD and aerosol optical parameters are calculated inline within the GFS v17 physics suite using the NASA GOCART look-up table, with aerosol input from either the diagnostic MERRA-2 climatological aerosol fields or the prognostic aerosol concentrations from the coupled aerosol prediction as in Cheng et al. (2026). This capability allows the prognostic aerosol optical properties derived from the inline-predicted aerosol mass concentrations and size distributions to directly influence shortwave and longwave radiative fluxes at each model time step, thereby representing aerosol radiative feedback that is absent in the one-way coupled configuration.

2.5 UFS-Chem model experiments

Experiments were performed using two configurations: (1) a cycling run, in which the atmosphere–aerosol model is initialized every 24-h with Global Data Assimilation System (GDAS) meteorological fields while aerosol fields are recycled from the previous cycle, representing an operational scenario and providing a near–best-case estimate of meteorological conditions for 1-day forecasts; and (2) a subseasonal run, a fully coupled configuration in which the atmosphere–aerosol system is coupled with the ocean, wave, and sea-ice components and initialized only once at the beginning to produce a 1-month prediction. Due to the unavailability of wave initial conditions for long-term simulations, the wave model was uncoupled for the 6-year experiments (2012–2017) to maintain consistency of each year. The atmosphere, ocean, and sea-ice components remained fully coupled; therefore, SST evolution was maintained through atmosphere–ocean coupling. The omission of wave coupling has a minor impact on the calculation of air–sea fluxes but is not expected to substantially affect the conclusions of this study. The details of UFS-Chem experiment configurations are described in Table 2 and 3. All subseasonal experiments are initialized using GDAS analyses for the meteorological variables and MERRA-2 reanalysis for the GOCART prognostic aerosol species and tracers. By isolating these two dimensions — feedback complexity and aerosol source — we aim to identify the dominant mechanisms through which interactive aerosol treatment improves subseasonal prediction fidelity. While control run uses radiative feedback from prescribed climatological aerosols, EXP1 is able to assess the impact of replacing MERRA-2 climatological aerosols with inline-predicted aerosols for radiative feedback, and EXP2 further includes aerosol indirect effects through interactive aerosol–cloud coupling using inline-predicted aerosols. Comparing Control and EXP1 isolates the effect of the aerosol radiative source (climatological versus inline-predicted) on forecast biases. Comparing EXP1 and EXP2 further isolates the incremental contribution of aerosol indirect effects, enabling attribution of bias reductions to aerosol–radiation and aerosol–cloud interaction pathways.

To assess the robustness of the single-case results from August 2016, we also extend the analysis to a 6-year period by conducting subseasonal experiments for each August from 2012 to 2017 in section 5. This study focuses on August, when biomass burning activity and aerosol loading are typically elevated, providing a suitable period for evaluating aerosol–radiation–cloud interactions. Although aerosol direct and indirect effects exhibit seasonal variability, extending the analysis to additional seasons requires substantially greater computational resources and is left for future work.

3. Improved performance of GEFS-Aerosols v12.3

To provide context for the development of the CCM-based GEFS-Aerosols (UFS-Chem), the following comparison summarizes the major improvements introduced in the operational GEFS-Aerosols v12.3 relative to GEFS-Aerosols v12. Since UFS-Chem is

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357 developed based on the GEFS-Aerosols v12.3 framework and these improvements have not been previously documented in the
 358 literature, this comparison in this section is included to establish the baseline capability of GEFS-Aerosols v12.3 rather than to
 359 directly evaluate UFS-Chem. The following experiments with both GEFS-Aerosol v12 and GEFS-Aerosol v12.3 are conducted in
 360 a cycling configuration consistent with operations. Figure 2 compares the monthly mean Day-1 AOD forecasts at 550 nm over
 361 North America for August 2021 from GEFS-Aerosols v12 and v12.3 against MERRA-2 reanalysis, the ICAP-MME, and satellite
 362 observations from MODIS and VIIRS. In GEFS-Aerosols v12, AOD values exceeding 1.0 are simulated over extensive regions of
 363 the western and central United States, producing a pronounced high bias that is not supported by the reanalysis, the multi-model
 364 ensemble, or the satellite retrievals. MERRA-2, ICAP-MME, MODIS, and VIIRS consistently indicate that elevated AOD during
 365 this period is largely confined to the Pacific Northwest and northern Great Plains, associated with wildfire smoke transport. GEFS-
 366 Aerosols v12.3 shows a substantial reduction in this positive AOD bias, with more spatially confined aerosol plumes and peak
 367 magnitudes that are in closer agreement with both the reanalysis products and satellite observations. The excessive westward and
 368 southward spread of high AOD values evident in v12 is notably reduced in v12.3, resulting in improved spatial distribution and
 369 more realistic AOD maxima. These improvements are primarily associated with the revised large-scale wet removal scheme and
 370 the corrected AOD calculation in v12.3, which together improve the representation of both the intensity and spatial extent of
 371 aerosol events over North America. OC is one of the dominant aerosol species emitted from wildfires. Comparison of OC AOD
 372 with the MERRA-2 reanalysis indicates that improved representation of OC AOD contributes substantially to the reduction of
 373 positive total AOD biases in GEFS-Aerosols v12.3 relative to v12 over North America. Figure 3 presents the spatial distribution
 374 of the monthly mean Day-1 forecast biases of total AOD and OC AOD for August 2021, defined as GEFS-Aerosols minus
 375 MERRA-2, for GEFS-Aerosols v12 and v12.3. In GEFS-Aerosols v12, pronounced positive total AOD biases are evident across
 376 much of western and central North America, with particularly large overestimations over the western United States and the
 377 downwind regions. The spatial pattern of OC AOD biases in v12 closely resembles that of the total AOD biases, indicating that
 378 the overestimation of organic aerosol loading is the major contributor to the high total AOD biases. In contrast, GEFS-Aerosols
 379 v12.3 shows a substantial reduction in both total AOD and OC AOD biases across large portions of North America, reflecting
 380 improvements associated with the model updates introduced in v12.3. Although the overall positive biases compared to MERRA-
 381 2 are largely reduced, weak negative total AOD biases emerge in some regions of the central and eastern United States. These
 382 negative biases against MERRA-2 likely reflect the combined effects of carbonaceous aerosols and sulfate aerosols. Overall, the
 383 comparison between v12 and v12.3 indicates that updates to fire emissions, wet deposition, and AOD calculations lead to
 384 substantial improvements in aerosol simulations across North America, providing a more robust aerosol baseline for the
 385 development of UFS-Chem.

386 In addition to evaluations against MERRA-2 reanalysis and satellite-based products, the performance of GEFS-Aerosols was
 387 further assessed using daily AERONET observations for August 2021. Table 4 summarizes the root mean square error (RMSE)
 388 and correlation coefficients of Day-1 AOD forecasts from GEFS-Aerosols v12, and v12.3 at approximately 25 AERONET sites
 389 located near major wildfire-affected regions across the United States and Canada. Corresponding scatter plots are shown in Fig.
 390 A1. The statistical results indicate substantial improvement in forecast performance in the updated v12.3 configuration. Correlation
 391 coefficients show notable improvement in v12.3, indicating better representation of day-to-day AOD variability. At PNNL, the
 392 correlation increases from 0.365 to 0.721, an improvement of approximately 97%. At Railroad Valley, the correlation improves
 393 from 0.395 to 0.732, an increase of approximately 85%, and at Neon Wood, from 0.482 to 0.751, an increase of approximately
 394 56%. These improvements in correlation are particularly significant at sites where wildfire-driven aerosol variability is most
 395 pronounced. For most sites over the western United States, RMSE values are also significantly reduced in v12.3 compared with
 396 v12. The most pronounced improvements are again observed at sites heavily influenced by wildfire smoke: at Bozeman, RMSE

decreases from 2.19 to 0.49, a reduction of approximately 78%; and at Rexburg Idaho, from 3.414 to 0.473, a reduction of approximately 86%. In California, similarly large reductions are evident at Bakersfield (from 1.19 to 0.288, approximately 76%), and NASA Ames (from 1.64 to 0.32, approximately 80%). The overall performance improvement is further illustrated in Fig. A1, which compares the correlation coefficients and RMSE values at individual AERONET sites between GEFS-Aerosols v12 and v12.3. Most sites lie above the 1:1 line for correlation and below the 1:1 line for RMSE, indicating that GEFS-Aerosols v12.3 generally improves AOD forecast skill by increasing correlation and reducing RMSE. Although GEFS-Aerosols v12.3 generally improves the AOD forecasts relative to v12, a few AERONET sites exhibit degraded performance. This likely reflects the regional response to multiple model updates, including the transition and updates of fire emissions and anthropogenic emissions, fixes to the dust emissions and AOD calculations, and improvements to wet deposition. These changes improve the overall forecast skill but may not produce uniform improvements at every individual site.

4. Implementing inline large-scale wet deposition into Thompson microphysics scheme for UFS-Chem

In many chemistry and aerosol modeling systems, large-scale wet removal is treated as an offline process, calculated within the chemistry or aerosol module using meteorological fields (e.g. large-scale precipitation etc.) provided by the host atmospheric model (Grell et al., 2005; Eastham et al., 2018; Appel et al., 2021; Collow et al., 2024). Because each chemistry or aerosol scheme applies its own internal logic for scavenging efficiency, vertical integration, and removal thresholds, this offline approach can produce scheme-dependent differences in aerosol removal — even when identical host model physics and precipitation fields are used as input. Such inconsistencies complicate model intercomparisons, reduce reproducibility, and can lead to divergent aerosol distributions and radiative effects across coupled configurations. To address this limitation, we implement large-scale wet removal inline within the Thompson microphysics scheme used in the GFSv17 physics suite, allowing aerosol scavenging to be handled directly by the physics that governs cloud formation and precipitation. This design is intended to improve consistency between aerosol removal and the host model’s cloud and precipitation processes, independent of the specific chemistry or aerosol mechanism employed. In addition, the inline implementation improves computational efficiency by avoiding the need to export meteorological fields to the chemistry or aerosol model for redundant wet-removal calculations. This framework provides a more consistent and extensible treatment of aerosol wet removal within UFS-Chem, facilitating robust application across cycling and fully coupled subseasonal configurations.

When the option for inline large-scale wet removal is enabled in the UFS model namelist, the Thompson microphysics scheme calculates large-scale wet removal of GOCART aerosol species through collection by precipitating hydrometeors. The coupling framework performs a mass-to-number conversion of prognostic GOCART aerosol species at each microphysics time step. The water-friendly aerosol number concentration (NWFA), representing cloud condensation nuclei (CCN), is calculated by summing contributions from sulfate, five sea salt bins, and hydrophilic carbonaceous species of hydrophilic OC. The ice-friendly aerosol number concentration (NIFA), representing ice-nucleating particles (INP), is derived from five dust bins (Cheng and Yang, 2025). Scavenging by rain, snow, and graupel is calculated during the microphysics integration using size-dependent collection efficiencies determined from the Stokes number, particle characteristics, and hydrometeor properties following Slinn (1983), Wang et al. (2010) and Thompson and Eidhammer (2014). The removal rates are computed as the product of an aerosol removal coefficient, collection efficiency, aerosol number concentration, and hydrometeor size-distribution parameters. Each aerosol species has a characteristic diameter used in the collection efficiency calculation, and the removal rate is proportional to the overlap between aerosol and hydrometeor size distributions. The wet-removed aerosol number concentrations are tracked separately for each species and precipitation type. The mass tendencies associated with this wet removal are communicated back to the GOCART

435 model at each physics time step through the chemistry–physics coupling interface. This maintains mass conservation and ensures
436 consistency between the microphysical scavenging and aerosol transport processes.

437 Figure 4 illustrates the impact of implementing inline large-scale wet removal within the Thompson microphysics scheme on
438 simulated AOD biases in both the 24-h cycling and fully coupled subseasonal configurations for August 2016. The model
439 configurations of these experiments are described in Table 2, and no aerosol indirect effect from inline predicted aerosol is enabled.

440 The CYC_OWR and CYC_IWR experiments represent the 24-h cycling runs with offline and inline large-scale wet removal,
441 respectively, whereas the SUB_OWR and SUB_IWR experiments represent the subseasonal free runs with offline and inline large-
442 scale wet removal, respectively. When the large-scale wet removal process is treated within the aerosol model of GEFS-GOCART
443 scheme, the CYC_OWR and SUB_OWR simulations exhibit markedly different AOD bias patterns, indicating strong sensitivity
444 to the free subseasonal run despite identical host-model physics. These discrepancies reflect inconsistencies between aerosol
445 removal and cloud–precipitation processes when wet scavenging is parameterized independently of atmospheric physics. In
446 practice, achieving relatively small AOD biases in the subseasonal configuration using the offline wet removal scheme tends to
447 worsen the performance in the cycling configuration (CYC_OWR), leading to larger biases there and highlighting the difficulty of
448 maintaining consistent aerosol behavior across forecast configurations. It should be noted that this behavior is not a universal issue
449 with offline wet-removal schemes, but rather reflects characteristics of the specific formulation used in GEFS-GOCART. Other
450 offline wet-removal parameterizations may exhibit better consistency across forecast configurations, depending on the
451 meteorological predictors used and their stability in subseasonal simulations. Using the implementation of inline large-scale wet
452 removal within the Thompson microphysics scheme, the AOD bias patterns become substantially more consistent between the
453 cycling (CYC_IWR) and subseasonal (SUB_IWR) simulations. Both configurations exhibit similar spatial structures and
454 comparable bias magnitudes, indicating that aerosol removal is now governed by the same cloud and precipitation processes across
455 timescales. This result highlights the advantage of implementing large-scale wet removal directly within the host model
456 microphysics scheme. By coupling aerosol scavenging to the same physical processes that produce clouds and precipitation, the
457 inline approach reduces configuration-dependent inconsistencies and provides a more robust framework when coupling different
458 aerosol or chemistry modules that may otherwise employ their own offline wet-removal parameterizations.

459 The divergence in AOD bias patterns between offline and inline wet-removal implementations reflects the complex sensitivity of
460 the offline scavenging formulation to multiple interacting model fields, which evolve differently in cycling versus subseasonal
461 configurations. The offline large-scale wet-removal scheme in the updated GEFS-GOCART calculates scavenging coefficient as
462 the ratio of precipitation rate to column-integrated cloud water (including density, vertical velocity, and layer thickness), applying
463 removal only where local cloud condensate exceeds a minimum threshold. This formulation introduces several pathways through
464 which systematic differences between cycling and subseasonal configurations can alter wet-removal efficiency. Even when bulk
465 precipitation amounts remain comparable, subseasonal runs may generate this precipitation through different microphysical
466 pathways—potentially involving reduced column-integrated cloud water, altered vertical velocity distributions, or more efficient
467 autoconversion processes. The layer-by-layer application of the threshold criterion means that configurations producing
468 comparable precipitation from fewer cloud-bearing layers will exhibit systematically reduced wet removal. Furthermore, the
469 denominator's dependence on instantaneous cloud water flux rather than accumulated precipitation creates a fundamental
470 inconsistency: aerosol scavenging becomes coupled to the model's diagnosed cloud state rather than to the precipitation that
471 physically drives the removal process. This dependence on instantaneous cloud state amplifies any systematic biases in cloud
472 microphysics that develop during extended free-running forecasts, providing a mechanistic explanation for the configuration-
473 dependent AOD bias divergence.

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475 In contrast, the inline Thompson implementation couples aerosol removal directly to hydrometeor collection processes within the
476 microphysics calculations. This approach reduces sensitivity to biases in diagnosed cloud condensate and ensures that aerosol
477 scavenging remains consistent with the precipitation physics across both cycling and subseasonal configurations. As a result, the
478 inline implementation provides a more physically consistent and robust treatment of large-scale wet removal within UFS-Chem.

479 5. ~~Implementation of aerosol indirect effect in UFS-Chem~~

480 Aerosol–cloud interactions are widely recognized as the most uncertain component of estimates of global radiative forcing
481 (Seinfeld et al., 2016). These interactions occur when aerosols act as CCN or INP, altering cloud microphysical properties such as
482 droplet number concentration and size distribution (IPCC, 2021). Such changes can subsequently influence precipitation processes
483 through cloud lifetime effects, whereby smaller droplets delay the onset of precipitation and may increase cloud spatial extent and
484 liquid water content (Seinfeld et al., 2016; Gordon et al., 2023; Li et al., 2024).

485 In the baseline Thompson microphysics scheme (Thompson et al., 2008), cloud droplet number concentration is prescribed as a
486 constant — 100 cm^{-3} over land and 50 cm^{-3} over ocean — thereby neglecting the spatiotemporal variability in aerosol
487 concentrations and their influence on cloud microphysical properties. Similarly, ice nucleation in the baseline Thompson
488 microphysics scheme relies on an empirical temperature-dependent parameterization (Cooper, 1986) rather than explicitly
489 representing INP populations, preventing the scheme from distinguishing between pristine and aerosol-laden environments.
490 Because cloud radiative properties and autoconversion rates (Berry and Reinhardt, 1974) are governed by these fixed assumptions,
491 the baseline configuration cannot represent aerosol indirect effects such as cloud albedo modification (Twomey, 1977) and
492 precipitation suppression (Albrecht, 1989). This lack of aerosol awareness is particularly limiting for extended-range forecasts, in
493 which episodic events such as dust outbreaks or wildfire (biomass burning) can significantly influence cloud properties and
494 precipitation efficiency.

495 Building upon the aerosol-aware Thompson–Eidhammer microphysics scheme (Thompson and Eidhammer, 2014), which utilizes
496 climatological aerosol fields as initial conditions and parameterized surface emissions to predict NWFA and NIFA that
497 subsequently act as CCN and INP, respectively, Li et al. (2024) developed an approach incorporating inline emission modules to
498 replace climatological emissions with more realistic and dynamic source terms. In this work, we develop and implement an option
499 within the GFSv17 physics suite of the UFS to enable fully prognostic aerosol–cloud interactions when coupled with the GEFS-
500 GOCART from GEFS-Aerosol v12.3. As illustrated in Fig. 5, this is achieved by coupling the GOCART aerosol model with the
501 Thompson microphysics scheme through a bidirectional framework that integrates inline large-scale wet removal processes. The
502 15 GOCART aerosol species are used within the coupling framework to support both aerosol–cloud interactions and inline wet
503 scavenging processes. The GOCART aerosol mass mixing ratios are converted to ice-friendly aerosol (IFA) and water-friendly
504 aerosol (WFA) number concentrations following Cheng et al. (2025). The IFA number concentration is derived from the five dust
505 size bins using the corresponding particle masses based on the bin median radius and particle density. The WFA number
506 concentration is computed from sea salt, sulfate, and hydrophilic organic carbon (OC2) using the prescribed conversion coefficients
507 of the Thompson aerosol-aware microphysics scheme. It should be noted that not all 15 species are mapped to activation proxies
508 for aerosol indirect effects. For the aerosol–cloud interaction component, only a subset of species is mapped to the NWFA and
509 NIFA proxies used by the microphysics activation parameterizations. The remaining aerosol species are not used in the CCN or
510 INP activation calculations but are still fully included in the inline large-scale wet removal processes. As a result, all 15 prognostic
511 GOCART aerosol species participate in the microphysics-based wet scavenging calculations, ensuring consistent aerosol removal
512 across the coupled system.

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Extending the work of Li et al. (2024) and Cheng et al. (2026), neither of which included inline aerosol–model coupling, this development refines the aerosol–microphysics coupling described in Section 4 and integrates inline wet scavenging within the Thompson microphysics scheme. This approach reduces coupling-related biases that can affect aerosol simulations in extended forecasts and enables a more consistent representation of aerosol–cloud interactions across weather and subseasonal prediction timescales. Increased aerosol loading leads to higher cloud droplet number concentrations and smaller mean droplet sizes. This change modifies cloud optical properties through the first indirect effect (Twomey, 1977) and suppresses the autoconversion of cloud water to rain through the second indirect effect (Albrecht, 1989), as illustrated in Fig. 5. In addition, dust aerosols act as INP and facilitate ice formation, with their concentration predicted using the empirical parameterization of DeMott et al. (2010) as a function of temperature and dust particle concentration. Higher concentrations of IFA increase ice crystal number concentrations and influence downstream ice microphysical processes, including depositional growth, aggregation, and riming, commonly referred to as the glaciation indirect effect.

A distinguishing feature of this implementation is the maintenance of physical consistency through mass-conserving aerosol budgets (Fig. 5). As described in Section 4, aerosol scavenging by precipitation is calculated inline within the microphysics scheme using size-dependent collection efficiencies. To maintain budget closure, aerosol activation into cloud droplets reduces NWFA proportionally to the diagnosed droplet number, ice nucleation consumes NIFA according to the activated ice crystal number, and wet removal decreases the concentrations of prognostic GOCART aerosol species according to the simulated hydrometeor collection rates. All aerosol tendencies derived from these processes are accumulated and returned to the chemistry component (Fig. 5). This feedback enables multi-day aerosol evolution within the coupled system, which is important for subseasonal prediction. Overall, this closed-loop coupling provides a unified framework in which aerosol emissions, chemistry, transport, wet and dry deposition, and cloud interactions are represented consistently within the host model physics, supporting applications that span weather to subseasonal prediction timescales.

6. Quantification of aerosol radiative ~~feedback~~ and aerosol indirect ~~effects~~ in subseasonal prediction

Aerosol–radiation and aerosol–cloud interactions represent key sources of uncertainty in S2S predictions. In many operational S2S systems, aerosol fields are prescribed from climatology, which neglects the radiative feedback of aerosol direct and semi-direct effects from prognostic aerosol variations and their indirect effects on cloud microphysics (Benedetti and Vitart, 2018; Bozzo et al., 2020; Fu et al., 2024).

To quantify these ~~aerosol–radiation and aerosol–cloud interaction effects~~, Figure 6 presents the total AOD biases relative to MERRA-2 reanalysis across three experiment configurations (Table 3), using August 2016 as a representative case study, a period that also corresponds to the evaluation of inline large-scale wet removal within the Thompson microphysics scheme (Fig. 4). All experiments use the inline large-scale wet removal, with differences arising from how aerosol radiative ~~feedback~~ and aerosol indirect ~~effects~~ are represented. In general, all three experiments show similar biases over broad regions, mostly within -0.1 to 0.1 (Fig. A2). The largest positive biases are mainly associated with wildfire regions over southern Africa and their downwind areas. In contrast, significant negative biases are primarily located over major dust source regions in northern Africa and the Middle East. The Control experiment, which employs MERRA-2 climatological aerosol fields for ~~radiative feedback of~~ direct and semi-direct ~~effects~~, exhibits substantial positive AOD biases over Northern Hemisphere high latitudes. Replacing the climatological fields with inline-predicted aerosols from GEFS-GOCART (EXP1) reduces these biases considerably at high latitudes. The further inclusion of aerosol indirect ~~effects~~ through ~~interactive~~ coupling with the Thompson aerosol-aware microphysics scheme (EXP2) yields additional bias reductions at high latitudes but introduces increased biases against MERRA-2 over the tropical Atlantic compared to the configuration using only radiative feedback (EXP1). These results suggest that transitioning from climatological to inline-

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560 predicted aerosols provides a substantial and widespread improvement in AOD representation, particularly at Northern Hemisphere
561 high latitudes, while the further inclusion of aerosol indirect effects introduces comparatively smaller but mixed regional effects.
562 To assess the robustness of the single-case results from August 2016, we extend the subseasonal analysis to a 6-year average of
563 each August from 2012 to 2017. Figure 7 presents the 6-year averaged 6-hourly evolution of total AOD and mean AOD bias with
564 respect to MERRA-2 reanalysis over forecast period for August in UFS-Chem (Control, EXP1, and EXP2), shown separately for
565 the tropical region (25°S–25°N) and the global average. The black line denotes the MERRA-2 reanalysis, which serves as the
566 verification reference. The grey line (MERRA-2 Clim) represents the AOD diagnosed from the MERRA-2 climatological aerosol
567 fields, which are derived from monthly mean MERRA-2 aerosol data averaged over 2003–2014. These climatological fields are
568 spatially interpolated onto the UFS grid and temporally interpolated at each model time step (Cheng and Yang, 2023). Aerosol
569 optical properties are then computed using the instantaneous relative humidity from the UFS forecast within the GFSv17 physics
570 suite. The aerosol optical properties diagnosed from the MERRA-2 climatological AOD are used to drive the radiative feedback
571 in the Control experiment. Both the tropical and global averages reveal that the MERRA-2 climatological AOD exhibits a persistent
572 negative bias of approximately -0.015 to -0.025 throughout the forecast period, substantially underestimating the aerosol loading
573 relative to the MERRA-2 reanalysis. In contrast, the three GOCART inline-predicted experiments produce AOD values that are
574 considerably more consistent with the MERRA-2 reanalysis, with substantially smaller biases. Both EXP1 and EXP2 use their
575 predicted aerosol fields to drive the radiative feedback. Among the three experiments, EXP2, which includes both aerosol radiative
576 feedback and aerosol indirect effects, exhibits the smallest biases over the full forecast period, followed by EXP1 with radiative
577 feedback only, and the Control without aerosol radiative feedback from the coupled aerosol model. This ordering is consistent
578 across both the tropical and global domains. A notable feature in the bias evolution is the significantly negative bias magnitude
579 during approximately the first 1–7 days of the forecast, followed by a gradual increase. This behavior reflects the aerosol spin-up
580 adjustment period: although the GOCART aerosol mixing ratios are initialized from MERRA-2 reanalysis, they are spatially
581 interpolated onto the UFS grid, and fundamental differences in the dynamical cores and physics parameterizations between the
582 two modeling systems introduce systematic discrepancies in the predicted AOD. These system-dependent differences, including
583 distinct treatments of aerosol sources and sinks, humidity, cloud processes, and radiative transfer, affect the evolution of aerosol
584 mixing ratios and their conversion to optical properties, resulting in the initial negative bias in AOD. As the forecast progresses
585 beyond the spin-up period, the GOCART aerosol fields adjust to the UFS dynamical and physical environment, and the predicted
586 AOD converges toward a more self-consistent state with reduced biases. After the spin-up period, the progressive divergence
587 among the experiments demonstrates the growing importance of aerosol-radiation and aerosol-cloud interaction effects, on
588 subseasonal timescales and confirms that the inclusion of aerosol indirect effects (EXP2) provides a consistent improvement in
589 AOD prediction throughout the extended forecast range.

590 In terms of spatial distribution, Figure 8 presents the 6-year averaged (2012–2017) total AOD biases with respect to MERRA-2
591 reanalysis for August. Compared with the representative single-year case of August 2016 (Fig. 6), the 6-year averaged AOD biases
592 are relatively larger and may also reflect the influence of interannual variability, which helps reveal the underlying systematic error
593 structure of each experiment more clearly. The spatial patterns of the largest positive and negative biases, associated with wildfire
594 and dust source regions, are broadly consistent with those observed in 2016 (Fig. 6). This consistency suggests that these regional
595 biases are more likely related to uncertainties in emission sources rather than interannual variability or aerosol feedback processes.
596 Meanwhile, the inter-experiment differences are smaller than those seen in the individual 2016 case (Fig. A2 and Fig. A3), which
597 may be attributable to interannual variability.

598 The Control experiment exhibits a global mean AOD bias of $+0.003$, with widespread positive biases against MERRA-2 over the
599 Northern Hemisphere high latitudes and central Africa, and negative biases concentrated over the southeastern United States, the

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504 Middle East, northern Africa, and South and East Asia. Replacing the climatological aerosol fields with inline-predicted aerosols
 505 from GEFS-GOCART (EXP1) shifts the global mean bias to +0.004, while reducing the positive biases against MERRA-2 at high
 506 latitudes near the North Pole compared with the Control experiment, but retaining the regional positive biases over major aerosol
 507 source regions. The addition of aerosol indirect ~~effects~~ (EXP2) reduces the global mean bias to +0.001, with modest improvements
 508 over high latitudes near the North Pole, the tropical Atlantic, and North America. Results across the 6-year period are consistent
 509 with the results from the single August 2016 case. The reduction in the global mean bias from +0.003 in the Control experiment
 510 and +0.004 in EXP1 to near-zero values in EXP2 indicates that the transition from climatological to inline-predicted aerosols with
 511 both ~~aerosol radiative feedback~~ and ~~aerosol indirect effects~~, substantially reduces the dominant systematic bias in AOD, whereas
 512 including aerosol radiative feedback alone does not significantly improve the AOD bias. These systematic improvements, which
 513 are largely masked by interannual noise in the single-year analysis, underscore the value of multi-year evaluation for quantifying
 514 the benefits of interactive aerosol treatment in subseasonal prediction systems. Figure 9 presents the 6-year averaged AOD RMSE
 515 with respect to MERRA-2 reanalysis for the three experiments. The Control experiment yields a global mean AOD RMSE of
 516 0.111. Replacing the climatological aerosol fields with inline-predicted aerosols from GEFS-GOCART for radiative feedback
 517 (EXP1) slightly increases the global mean RMSE to 0.113, although notable improvements are observed over eastern Europe and
 518 Russia. The further inclusion of aerosol indirect ~~effects~~ (EXP2) reduces the global mean RMSE to 0.111 relative to EXP1, with
 519 the most pronounced improvements occurring over North America. The overall reduction in AOD RMSE when both ~~aerosol~~
 520 radiative ~~feedback~~ and ~~aerosol indirect effects~~ are included, particularly at regional scales confirms that the progressive activation
 521 of aerosol radiative ~~feedback~~ and ~~aerosol indirect effects~~ can improve aerosol prediction fidelity in UFS-Chem on subseasonal
 522 timescales.

523 Beyond the improvements in AOD representation, the progressive activation of ~~aerosol radiative feedback and aerosol indirect~~
 524 ~~effects~~ also influences key meteorological fields relevant to subseasonal prediction. Figure 10 presents the 6-year averaged (2012–
 525 2017) net radiative flux bias at the top of the atmosphere (TOA) for August in UFS-Chem, evaluated against CERES observations.
 526 The Control experiment, which uses MERRA-2 climatological aerosols for radiative feedback, exhibits a global mean TOA net
 527 radiative flux bias of -3.24 W m^{-2} , with widespread negative biases over tropical and mid-latitude oceans indicating excessive
 528 outgoing radiation, and positive biases over parts of the Southern Ocean, the southeastern Pacific, and high-latitude regions.
 529 Replacing the climatological aerosol fields with inline-predicted aerosols from GEFS-GOCART for radiative feedback (EXP1)
 530 increases the magnitude of the global mean bias to -3.56 W m^{-2} , with enhanced negative biases over the tropical oceans and the
 531 North Atlantic. This degradation indicates that the radiative effects associated with inline-predicted aerosols are not fully balanced
 532 by other components of the model physics. The further inclusion of aerosol indirect ~~effects~~ (EXP2) substantially reduces the global
 533 mean TOA net radiative flux bias to -2.23 W m^{-2} , representing a 37% improvement relative to EXP1 and a 31% improvement
 534 relative to the Control. Spatially, the bias patterns across the three experiments are broadly similar, although EXP2 shows a notable
 535 reduction in negative biases over the Maritime Continent and surrounding regions. These results indicate that while the ~~radiative~~
 536 ~~feedback of~~ direct and semi-direct ~~effects~~ of inline-predicted aerosols alone can slightly degrade the TOA radiation balance, the
 537 inclusion of aerosol–cloud microphysical coupling helps reduce the global TOA energy imbalance.

538 Figure 11 presents the 6-year averaged (2012–2017) cloud fraction biases against CERES observation for August across the three
 539 experiments. The Control experiment, which relies on MERRA-2 climatological aerosols for direct and semi-direct ~~effects~~, exhibits
 540 a global mean cloud fraction bias of -4.57% , indicating a systematic underestimation of cloudiness. Replacing the climatological
 541 fields with inline-predicted aerosols from GEFS-GOCART (EXP1) reduces the global mean cloud fraction bias to -3.98% ,
 542 representing an approximately 13% improvement. However, the spatial patterns of cloud fraction bias remain remarkably similar
 543 between the Control and EXP1, with the largest negative biases concentrated off the western coasts of North and South America

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— regions that do not correspond to the areas of largest AOD bias or RMSE in Fig. 9. This suggests that the modest global mean improvement in cloud fraction between Control and EXP1 is not straightforwardly attributable to regional aerosol–radiation interactions, and that the dominant sources of cloud fraction bias are likely governed by other model physics, such as boundary layer parameterization and large-scale dynamics, rather than aerosol loading alone. The further inclusion of aerosol indirect effects through the Thompson aerosol-aware microphysics scheme (EXP2) achieves an additional substantial reduction in cloud fraction bias to -2.87% , corresponding to an approximately 28% further improvement relative to EXP1. This additional gain is most pronounced over the North Atlantic, where the coupling between aerosol concentrations and cloud droplet number concentration directly modifies cloud microphysical properties and lifetime. The interactive aerosol–cloud coupling pathway—through which increased aerosol loading enhances droplet number concentrations, suppresses autoconversion, and prolongs cloud lifetime—introduces microphysical processes that are not represented by radiative feedback of direct and semi-direct effects alone. Consequently, the substantial reduction in systematic AOD biases from EXP1 to EXP2 enables a more realistic representation of aerosol–cloud interactions, leading to larger adjustments in cloud properties, including cloud fraction, and contributing to improvements in the TOA radiation balance.

The SST biases (Fig. 12) compared to OISST further illustrate the downstream impact of aerosol radiative feedback and aerosol indirect effects on the coupled ocean–atmosphere system. The Control and EXP1 experiments produce identical global mean SST biases of -0.11 K and -0.12 K, indicating that the transition from climatological to inline-predicted aerosols for direct and semi-direct effects alone does not substantially alter the surface energy balance on subseasonal timescales. However, the inclusion of aerosol indirect effects in EXP2 reduces the global mean SST bias to -0.08 K, with the most notable warming corrections occurring over the North Pacific Ocean. This SST improvement is consistent with the enhanced cloud fraction in EXP2: the aerosol indirect effects increases cloud cover predominantly in regions where the Control and EXP1 experiments underestimate cloudiness, thereby modifying the surface shortwave radiation balance and reducing the cold SST bias. The coherent improvements across AOD, cloud fraction, and SST from Control through EXP1 to EXP2 demonstrate that the benefits of interactive aerosol treatment propagate through the aerosol–radiation–cloud–ocean coupling chain, underscoring the importance of including aerosol radiative feedback and aerosol indirect effects for subseasonal prediction fidelity.

Figure 13 presents the 6-year averaged (2012–2017) differences in total, large-scale and convective precipitation for August, decomposed into the contributions from aerosol radiative feedback, aerosol indirect effects, and their combination. Enabling aerosol radiative feedback from inline-predicted GOCART aerosols instead of MERRA-2 climatological aerosols (EXP1 minus Control; left column) produces only small and spatially scattered changes in total precipitation, with modest increases over the tropical western Pacific and the Maritime Continent and slight reductions over parts of the tropical Indian Ocean and eastern Pacific. At the process level, changes in large-scale precipitation are minimal and spatially incoherent, while convective precipitation shows the strongest signal over the tropical Indian Ocean through the Maritime Continent to the tropical western Pacific. The differences in aerosol loading between the inline-predicted and climatological fields alter atmospheric shortwave absorption and scattering profiles, modifying atmospheric heating rates, stability, cloud formation, and the large-scale circulation patterns that govern precipitation distribution.

Further enabling the aerosol indirect effects (EXP2 minus EXP1; middle column of Fig. 13) produces more pronounced and spatially coherent changes in total precipitation, with widespread enhancements over the tropics, the Maritime Continent, and southeastern Asia, and notable reductions over the tropical central and eastern Pacific and parts of the Southern Ocean (global mean difference of $+0.04$ mm day $^{-1}$). At the process level, large-scale precipitation shows widespread enhancements over the tropics and southeastern Asia, with reductions over the tropical central Pacific and parts of the Southern Ocean (global average of -0.01 mm

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703 day⁻¹), while convective precipitation drives substantial increases over the tropical western Pacific and the Maritime Continent,
704 with notable decreases over the tropical eastern Pacific and the Indian Ocean (global average of +0.05 mm day⁻¹).

705 The combined effect of radiative ~~feedback and aerosol indirect effects~~, (EXP2 minus Control; right column) reveals a total
706 precipitation response that is larger and more spatially organized than the radiative feedback alone, with enhanced precipitation
707 concentrated over the tropical western Pacific, the Maritime Continent, and southeastern Asia, and reduced precipitation over the
708 tropical eastern Pacific, parts of the Indian Ocean, and the Southern Ocean (global mean difference of +0.04 mm day⁻¹). At the
709 process level, increased large-scale precipitation is evident over the tropics and southeastern Asia, with reductions over the
710 Southern Ocean (global average of -0.02 mm day⁻¹), while convective precipitation increases are concentrated over the tropical
711 western Pacific and the Maritime Continent, with decreases over the tropical eastern Pacific and the Indian Ocean (global average
712 of +0.06 mm day⁻¹). Notably, precipitation differences in EXP2 minus Control are generally larger and more spatially coherent
713 than those in EXP1 minus Control, particularly for convective precipitation, whereas the response in large-scale precipitation
714 remains relatively weak. This amplification likely occurs because the ~~aerosol indirect effects~~ influences cloud microphysical
715 properties by increasing cloud droplet number concentration, reducing autoconversion efficiency, and prolonging cloud lifetime.
716 These changes can subsequently affect latent heat release, atmospheric stability, and large-scale circulation, leading to adjustments
717 in the distribution of precipitation. The radiative feedback, by contrast, primarily modulates the thermodynamic profile through
718 changes in shortwave absorption and longwave emission, producing a comparatively more modest precipitation response. The
719 spatial coherence between the combined precipitation response and the improvements in AOD (Figs. 8 and 9), cloud fraction, and
720 SST (Fig. 12) demonstrates that the benefits of interactive aerosol treatment propagate through the aerosol–radiation–cloud–
721 precipitation–ocean coupling chain, with the ~~aerosol indirect effects~~ playing the dominant role in reshaping the convective
722 precipitation distribution in subseasonal predictions.

723 However, the enhanced convective precipitation does not simply coincide spatially with the regions of reduced biases for net
724 radiative flux at the TOA, cloud fraction and SST, indicating that the ~~aerosol indirect effects~~ modifies the precipitation response
725 through complex interactions between cloud microphysical changes and large-scale dynamical adjustments. In regions where the
726 suppression of autoconversion extends cloud lifetime and increases cloud water content, the eventual release of accumulated
727 moisture can enhance convective activity, while in other regions the stabilization of the lower troposphere through increased
728 cloudiness may redistribute convective precipitation spatially.

729 7. Summary and discussion

730 The development of the CCpp-based GEFS-Aerosols framework within UFS-Chem represents a significant advancement toward
731 a more physically consistent and tightly coupled atmospheric composition modeling system. By embedding aerosol and chemistry
732 processes directly within the atmospheric model physics time step, the CCpp architecture allows vertical tracer transport, mixing,
733 scavenging, deposition, and aerosol–radiation interactions to be handled using the same physical parameterizations that govern
734 meteorological processes. This unified framework reduces inconsistencies inherent in the traditional NUOPC-style separation
735 between physics and chemistry components and avoids redundant calculations by leveraging meteorological fields already
736 available within the host physics suite, thereby improving both physical consistency and computational efficiency. Within this
737 framework, GEFS-GOCART from GEFS-Aerosols v12.3 is adopted as the baseline aerosol module, incorporating updated
738 anthropogenic emissions, revised large-scale wet removal parameterizations, and corrected AOD calculations. Evaluation against
739 MERRA-2 reanalysis, satellite retrievals (MODIS and VIIRS), and AERONET observations shows that these updates substantially
740 reduce the high AOD biases present in the earlier v12 configuration, particularly over wildfire-affected regions of western North
741 America, largely due to improved representation of organic carbon aerosols and more realistic wet removal processes.

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746 To further improve the physical consistency of aerosol–cloud–precipitation interactions, this study implements inline large-scale
747 wet removal within the Thompson microphysics scheme. Unlike traditional offline approaches that diagnose aerosol removal from
748 exported precipitation and cloud fields, the inline implementation couples aerosol scavenging directly with hydrometeor formation
749 and precipitation processes within the microphysics integration. It eliminates redundant diagnostic calculations and ensures that
750 aerosol removal is governed by the same physical processes responsible for cloud and precipitation formation. Model experiments
751 show that this inline treatment substantially improves the consistency of simulated AOD biases between cycling and fully coupled
752 subseasonal configurations, indicating reduced sensitivity of aerosol removal to configuration-dependent cloud water biases. It
753 should be noted that this inconsistency is not a universal limitation of offline wet-removal schemes, but rather reflects
754 characteristics of the specific large-scale wet-removal formulation used here. Other offline parameterizations may exhibit better
755 consistency across forecast configurations, depending on the meteorological predictors used and their stability in subseasonal
756 simulations. These results highlight the advantage of implementing large-scale wet removal directly within the host model
757 microphysics scheme.

758 In addition, aerosol indirect effects are implemented through inline coupling between the prognostic GOCART aerosol model and
759 the Thompson microphysics scheme. In contrast to the baseline configuration that prescribes fixed cloud droplet concentrations,
760 the new implementation enables aerosol-aware microphysics by mapping GOCART aerosol species to water-friendly and ice-
761 friendly aerosol populations that act as CCN and INP, respectively. Cloud droplet and ice crystal number concentrations are then
762 computed prognostically based on aerosol loading and environmental conditions, allowing aerosols to modify cloud optical
763 properties and precipitation processes through established indirect effects. Together with the implementation of two-way aerosol–
764 radiation coupling in UFS-Chem, which allows prognostic aerosol optical properties to directly influence radiative fluxes, this
765 framework provides a unified and physically consistent representation of aerosol–radiation–cloud interactions. These
766 developments establish a robust and extensible aerosol modeling framework within UFS-Chem that is suitable for applications
767 spanning weather forecasting to S2S prediction and future Earth system modeling.

768 Sensitivity experiments demonstrate that progressively activating aerosol radiative ~~feedback and aerosol indirect effects~~ in UFS-
769 Chem influences the simulation of aerosols, clouds, radiation, and precipitation on subseasonal timescales. The multi-year
770 evaluation shows that replacing climatological aerosols with inline-predicted aerosols alone does not significantly reduce AOD
771 bias and can slightly increase global AOD RMSE. However, when aerosol indirect effects are included, the global mean AOD bias
772 is reduced to near-zero values and regional RMSE improvements become more evident, particularly over North America. These
773 results indicate that aerosol–cloud microphysical interactions play a key role in improving aerosol prediction fidelity in UFS-
774 Chem, while radiative feedback alone provides limited benefit for correcting systematic AOD errors. Despite these improvements
775 in aerosol representation, the results also highlight an important challenge: more realistic aerosol treatments do not lead to uniform
776 improvements across variables and regions. For example, although the inclusion of interactive aerosols improves cloud fraction
777 and reduces AOD biases in several regions, the radiative feedback alone slightly worsens the TOA net radiative flux bias, and the
778 inclusion of aerosol indirect effects can increase AOD biases over the tropical Atlantic even as cloud properties improve elsewhere.
779 Although the inclusion of aerosol indirect effects improves cloud fraction, TOA radiative fluxes, and SST, the present analysis
780 does not isolate whether these improvements result from a more realistic representation of aerosol–cloud interactions or from a
781 different balance of compensating model errors. Additional evaluations using cloud microphysical properties and process-oriented
782 diagnostics will be needed to further assess the underlying mechanisms. The precipitation response further illustrates this
783 complexity. Enhanced convective precipitation does not spatially coincide with regions of improved cloud fraction or reduced
784 TOA radiation biases, and large-scale precipitation exhibits a net global reduction despite improvements in aerosol and cloud
785 representations. Notably, the magnitude of precipitation differences in EXP2 minus Control is substantially larger than in EXP1

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787 minus Control for total precipitation, which is dominated by convective components, indicating that aerosol indirect effects amplify
788 the precipitation response beyond what radiative feedback alone can produce. These responses likely reflect the fact that the current
789 model physics were tuned to maintain approximate energy balance under climatological aerosol forcing. Introducing more realistic,
790 time-varying aerosol fields perturbs this balance and can expose compensating errors in other components of the model physics.
791 Fully realizing the benefits of interactive aerosol treatment for subseasonal prediction may therefore require coordinated retuning
792 and improved representation of related processes, including radiation, cloud microphysics, convection, and surface energy
793 exchange.

794 It should be noted that the CCpp-based GEFS-Aerosols implementation is evaluated within the complete UFS-Chem framework.
795 Therefore, the reported performance reflects the combined effects of the UFS-Chem configuration, and the individual contributions
796 of the tight physics–aerosol coupling and other model updates are not isolated. Although some biases remain in UFS-Chem for
797 subseasonal prediction, substantial progress has been made toward improving global aerosol and chemistry simulations within the
798 UFS framework.

799 Looking ahead, several developments are planned to further enhance the representation of aerosol–radiation–cloud interactions in
800 UFS-Chem v2. Several aerosol species are absent from the present configuration. The bulk GOCART module in GEFS-Aerosols
801 carries no secondary organic aerosol, nitrate, or ammonium, so these contributions are missing from the aerosol fields used here.
802 Volcanic emissions are supported by our configuration but were not activated, as no significant eruption occurred during the
803 simulation periods. Including these species is planned for future development, and applying the same framework to a volcanically
804 perturbed period is a natural extension, given that prognostic volcanic emissions substantially alter simulated aerosol burden and
805 optical properties (Mills et al., 2016) and that the resulting sulfate size distribution is sensitive to the treatment of chemistry and
806 aerosol modes (Hu et al., 2025). Although several aerosol processes have been tightly coupled within the GFSv17 physics suite
807 using CCpp, the dry deposition process remains decoupled from the GFSv17 PBL scheme, as GEFS-GOCART currently uses an
808 independent PBL mixing scheme for this purpose. Fully coupling dry deposition with the GFSv17 PBL scheme represents a
809 planned future development. In the current UFS configuration, several radiatively important gaseous tracers, including CO₂ and
810 O₃, are prescribed from external datasets or represented using simplified chemistry, which introduces additional uncertainty in the
811 simulated radiative balance. In future development of CATChem v2, these key radiatively active species will be predicted by the
812 comprehensive AM4.1 gas-phase chemistry framework (He et al., 2026) with interactive stratospheric chemistry, rather than
813 remaining prescribed as in the current version. Replacing prescribed radiatively important tracers with fully coupled inline
814 chemistry is expected to improve the consistency between atmospheric composition and radiation calculations and reduce
815 uncertainties in aerosol–radiation feedbacks. The current UFS-Chem framework employs the GOCART aerosol module, which
816 uses a bulk single-moment approach that tracks aerosol mass concentrations but does not explicitly predict aerosol number
817 concentrations. While computationally efficient, this scheme imposes inherent limitations on the representation of aerosol size
818 distributions and aerosol–cloud interactions, as the activation of aerosol particles into cloud droplets cannot be physically
819 constrained without prognostic number concentrations, leading to uncertainties in cloud radiative forcing. To address these
820 limitations, CATChem v2 will introduce a two-moment aerosol scheme that explicitly predicts both aerosol mass and number
821 concentrations, providing a more physically realistic representation of aerosol size distributions and is expected to improve the
822 simulation of cloud droplet activation, precipitation processes, and aerosol indirect radiative effects.

823 The present study evaluates the impacts of aerosol indirect effects through cloud fraction, top-of-atmosphere radiative fluxes, SST,
824 and cloud liquid water path. The cloud liquid water path comparison further demonstrates enhanced cloud water associated with
825 the inclusion of aerosol indirect effects (Fig. A4), consistent with the findings of the previous study Li et al. (2024). A more
826 comprehensive evaluation of aerosol indirect effects would benefit from additional cloud microphysical diagnostics, such as cloud

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327 droplet effective radius, together with comparisons against corresponding observations. As this work represents the first
328 implementation of inline aerosol indirect effects within the tightly coupled UFS-Chem framework, these diagnostics are not yet
329 available in the current system. Expanding the diagnostic output and conducting a comprehensive observational evaluation of
330 aerosol indirect effects will be a focus of future UFS-Chem development. Together, these developments will help establish a more
331 comprehensive Earth system modeling capability within UFS-Chem and support future improvements in S2S prediction.

332

333 Appendices

334 Appendix A

335 Figure A1 indicates the scatter comparison of Day-1 AOD forecast performance at approximately 25 AERONET sites over the
336 United States and Canada for August 2021. Figure A2 and A3 show the spatial differences in total AOD for August in UFS-
337 Chem, isolating the aerosol radiative ~~feedback and aerosol indirect effects~~ across the three experimental configurations. Figure
338 A4 shows the cloud liquid water path. Table A1 is the list of acronyms, abbreviations, and experiment names used in this study.

339 Code and data availability

340 The current version of CATChem is available from the project repository at <https://github.com/ufs-community/CATChem> under
341 the Apache-2.0 license. The versions of GEFS-Aerosol v12.3 and UFS-Chem v1 used to produce the results presented in this paper
342 are archived on Zenodo under DOI <https://doi.org/10.5281/zenodo.19262140> (Zhang et al., 2026).

343 Author contributions

344 LZ prepared the manuscript with contributions from all co-authors, she was one of the major developers of GEFS-Aerosols and
345 the lead developer of the CCPP-based GEFS-Aerosols (the aerosol component of UFS-Chem v1), including coupling GEFS-
346 GOCART to the UFS/GFSv17 using the CCPP, implementing inline large-scale wet removal and aerosol indirect ~~effects~~ through
347 inline-coupled GEFS-GOCART, and developing the aerosol-related components within the global workflow. HL was one of the
348 major developers of the initial CCPP coupling between GEFS-GOCART and FV3/GFSv15. GAG proposed the original concept
349 of tight coupling between aerosol/chemistry and physics and provided oversight of model development for both GEFS-Aerosols
350 and UFS-Chem. PSB performed model evaluation for GEFS-Aerosols and UFS-Chem. GAF performed data analysis and model
351 evaluation for UFS-Chem. BWG downloaded and post-processed the CERES and OISST datasets and processed model output
352 used in Figs. 10–13. SS provided suggestions for subseasonal studies. LB provided supervision and contributed to the organization
353 of the manuscript. AJ provided suggestions for inline large-scale wet removal and aerosol indirect ~~effects~~ in the Thompson
354 microphysics scheme. BB is the lead developer of the FENGSHA dust scheme. BB and LP assisted with updates and the operational
355 transition of GEFS-Aerosols v12.3. JH assisted with the reconstruction of CATChem v1. JS provided suggestions for implementing
356 inline large-scale wet removal. RA provided suggestions for plume-rise module optimization and supervision. ST assisted with
357 plume-rise module optimization and model debugging. DS provided assistance and suggestions for CCPP coupling. AC and FY
358 provided suggestions for aerosol indirect ~~effects~~ in the Thompson microphysics scheme; FY also provided supervision for the
359 operational transition of GEFS-Aerosols v12.3. RHS and BCM provided suggestions and project management for UFS-Chem. SK
360 developed the GBBEPx v4 dataset. DH assisted with CCPP-related code management during the early stages of development. HL,
361 GAG, PSB, GAF, BWG, SS, LB, AJ, JH, DS, RHS, and BCM provided suggestions to improve the manuscript.

362 Competing interests

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367 The authors declare that they have no conflict of interest.

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375 **Disclaimer**

376 The statements, findings, conclusions, and recommendations are those of the author(s) and do not necessarily reflect the views of
377 NOAA or the U.S. Department of Commerce.

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127 **Table 1.** Linkages and comparison among the global aerosol forecast systems: GEFS-Aerosols v12, GEFS-Aerosols v12.3, and
128 UFS-Chem.

Model name	GEFS-Aerosols v12	GEFS-Aerosols v12.3	UFS-Chem
Operational status	September 2020 - January 2023	Since January 2023	No
Dynamic core and resolution	FV3 (~25km, L65)	FV3 (~25km, L65)	FV3 (~25km, L127)
Physics	GFSv15	GFSv15	GFSv17
Infrastructure	NUOPC	NUOPC	CCPP
Aerosol model	GEFS-GOCART	Updated GEFS-GOCART (see section 2.2)	Based on GEFS-Aerosols v12.3 with updated FENGSHA dust input (see section 2.4)
Anthropogenic emission	CEDS v2014	CEDS v2019	CEDS v2019
Biomass burning emission	GBBEPx v3	GBBEPx v4	GBBEPx v4
Option to include inline aerosol radiative feedback	No	No	Yes
Option to include aerosol indirect effects _v	No	No	Yes
Option to couple with other components for S2S prediction	No	No	Yes

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140 **Table 2.** UFS-Chem experiments for large-scale wet removal.

Experiment	CYC OWR	CYC IWR	SUB OWR	SUB IWR
Component	ATM+Aero+Land	ATM+Aero+Land	ATM+Aero+Land+Ocean+seai+wave	ATM+Aero+Land+Ocean+seai+wave
Large-scale wet removal	Offline	Inline	Offline	Inline
Aerosol for radiative feedback	MERRA-2 climatology	MERRA-2 climatology	Inline updated GEFS-GOCART	Inline updated GEFS-GOCART
Simulation	Cycling run	Cycling run	Subseasonal free run	Subseasonal free run

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Table 3. UFS-Chem subseasonal experiments for aerosol radiative feedback and aerosol indirect effects.

Experiment	Control	EXP1	EXP2
Aerosol source for radiative feedback	MERRA-2 climatology	Inline updated GEFS-GOCART	Inline updated GEFS-GOCART
Aerosol indirect effects from inline aerosol prediction	Off	Off	On

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153 **Table 4.** AERONET site information, the correlation coefficients and RMSE of GEFS-Aerosols v12, GEFS-Aerosols v12.3 and
154 MERRA-2 AOD with respect to that of AERONET observation for the period 8/1/2021-8/31/2021. The correlation coefficients
155 are statistically significant at the 95% confidence level.

Station Names	(Latitude, Longitude)	Corr. with GEFS-Aerosols v12	Corr. with GEFS-Aerosols v12.3	RMSE with GEFS-Aerosols v12	RMSE with GEFS-Aerosols v12.3
U. of Reno	(39.54N, 119.81W)	0.601	0.546	2.44	0.844
Neon_Wref	(45.82N, 121.95W)	0.8425	0.858	0.314	0.348
PNNL	(46.34N, 119.28W)	0.365	0.721	1.282	0.455
Meridian	(43.60N, 116.35W)	0.599	0.724	0.77	0.552
Cascade Airport	(44.50N, 116.02W)	0.498	0.575	0.65	0.63
Taylor Ranch	(45.10N, 114.85W)	0.411	0.686	1.6	0.66
Lewis Clark	(46.41N, 117.03W)	0.420	0.56	1.14	0.64
Pinehurst	(47.54N, 116.24W)	0.507	0.69	0.769	0.602
Missoula	(46.92N, 114.08W)	0.610	0.675	0.541	0.455
Bozeman	(45.66N, 111.05W)	0.588	0.677	2.19	0.4898
Rexburg Idaho	(43.82N, 111.78W)	0.493	0.697	3.414	0.473
Bakersfield	(35.33N, 119.00W)	0.7299	0.734	1.199	0.288
Monterey	(36.59N, 121.85W)	0.814	0.847	1.52	0.4
NASA AMES	(37.42N, 122.06W)	0.863	0.828	1.64	0.32
Railroad Valley	(38.50N, 115.69W)	0.395	0.732	0.954	0.418
Neon Wood	(47.13N, 99.24W)	0.482	0.751	0.643	0.336
U. of Wisconsin	(43.07N, 89.41W)	0.760	0.64	0.28	0.213
GSFC	(38.99N, 76.84W)	0.485	0.575	0.172	0.12
Cartel (NY)	(45.38N, 71.93W)	0.648	0.69	0.255	0.253
Toronto	(43.79N, 79.47W)	0.45	0.55	0.311	0.179
Appalachian	(36.21N, 81.69W)	0.737	0.719	0.097	0.097
Tucson	(32.23N, 110.95W)	0.52	0.54	0.05	0.05

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Figure Captions.

Figure 1. Schematic comparison of NUOPC-based coupling in GEFS-Aerosols (top) and the CCPP-based implementation in UFS-Chem (bottom), highlighting the transition from externally coupled chemistry to chemistry and aerosol processes embedded directly within the FV3 physics suite. Color coding indicates process origin: orange denotes original GOCART aerosol processes; blue indicates aerosol-related processes inline-merged into the physics suite; and green represents original physics processes. The box is labelled "GOCART Aerosol/Chemistry" as interactive gas-phase chemistry (AM4.1) is included alongside the GOCART aerosol module.

Figure 2. Monthly mean Day-1 AOD forecasts for August 2021 over North America from (a) GEFS-Aerosols v12.3, (b) GEFS-Aerosols v12 compared with (c) MERRA-2 reanalysis; (d) MODIS; (e) ICAP multi-model ensemble; and (f) VIIRS observation. All panels show AOD at 550 nm.

Figure 3. Monthly mean Day-1 Total AOD (top) and OC AOD (bottom) forecast biases for August 2021, defined as GEFS-Aerosols minus MERRA-2, for GEFS-Aerosols v12 (left) and v12.3 (right).

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Figure 5. Schematic of the GOCART–Thompson coupling framework for prognostic aerosol–cloud interactions in UFS-Chem. NIFA and NWFA are the ice-friendly aerosol number concentration and water-friendly aerosol number concentration, respectively.

Figure 6. AOD biases relative to MERRA-2 reanalysis for August 2016 in UFS-Chem. (a) Control: aerosol radiative feedback from prescribed MERRA-2 climatological aerosol fields; (b) EXP1: aerosol radiative feedback from inline-predicted aerosols; (c) EXP2: aerosol radiative feedback and aerosol indirect effects from inline-predicted aerosols.

Figure 7. Six-year averaged (2012–2017) 6-hourly total aerosol optical depth (AOD) and mean AOD bias with respect to MERRA-2 reanalysis forecast period for August in UFS-Chem. Top row: tropical region average (25°S–25°N); bottom row: global average. (a, c): daily average total AOD; (b, d): mean bias. The black line denotes the MERRA-2 reanalysis. The grey line represents AOD diagnosed from MERRA-2 climatological aerosol. The blue, red, and green lines represent inline-predicted AOD from the Control, EXP1, and EXP2 experiments, respectively.

Figure 8. The same as in Figure 6, but averaged over the six-year of August (2012–2017).

Figure 9. The same as in Figure 8, but for AOD RMSE.

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Figure 13. Six-year averaged (2012–2017) differences in total precipitation (top row; mm day⁻¹), large-scale precipitation (middle row; mm day⁻¹) and convective precipitation (bottom row; mm day⁻¹) for August in UFS-Chem. (a) Effect of aerosol radiative feedback (EXP1 minus Control). (b) Aerosol indirect effects (EXP2 minus EXP1). (c) Combined effect of aerosol radiative feedback and aerosol indirect effects (EXP2 minus Control).

Figure A1. Scatter comparison of Day-1 AOD forecast performance at approximately 25 AERONET sites over the United States and Canada for August 2021. (a) Correlation coefficient and (b) root mean square error (RMSE) for GEFS-Aerosols v12.3 versus GEFS-Aerosols v12. The solid diagonal line denotes equal performance between the two model versions.

Figure A2. Differences in total AOD for August 2016 in UFS-Chem. (a) Effect of aerosol radiative feedback (EXP1 minus Control). (b) Aerosol indirect effects (EXP2 minus EXP1). (c) Combined effect of aerosol radiative feedback and aerosol indirect effects (EXP2 minus Control).

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236 **Figure A3.** Six-year averaged (2012–2017) differences in total AOD for August in UFS-Chem. (a) Effect of aerosol radiative
237 feedback (EXP1 minus Control). (b) Aerosol indirect effects (EXP2 minus EXP1). (c) Combined effect of aerosol radiative
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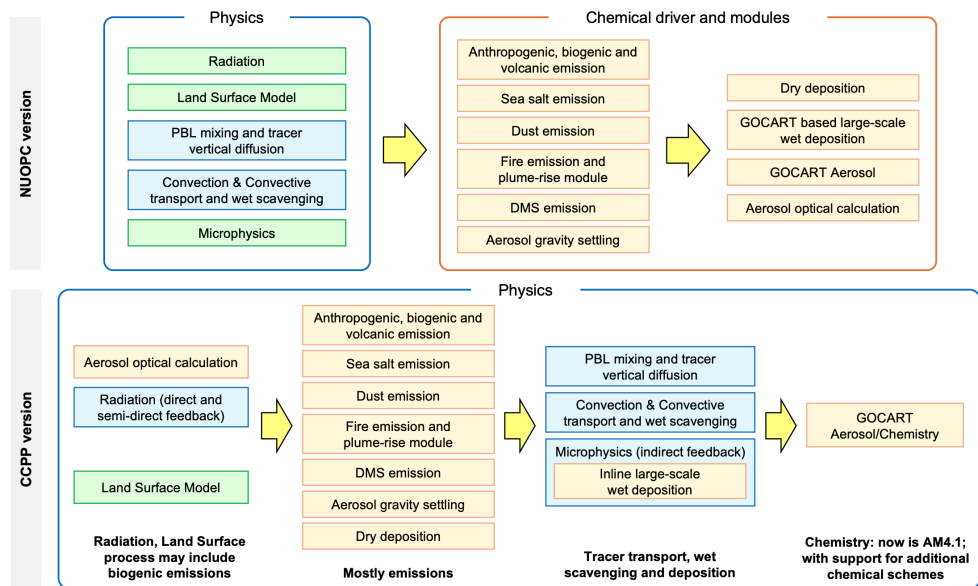
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240 **Figure A4.** Cloud liquid water path for August 2016 in UFS-Chem. (a) Control: aerosol radiative feedback from prescribed
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244 **Table A1.** List of acronyms, abbreviations, and experiment names used in this study.

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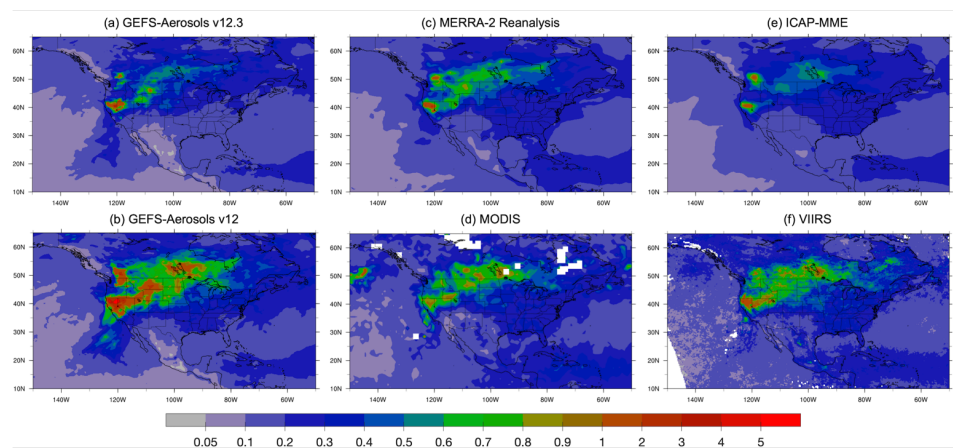
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Figure 2. Monthly mean Day-1 AOD forecasts for August 2021 over North America from (a) GEFS-Aerosols v12.3, (b) GEFS-Aerosols v12 compared with (c) MERRA-2 reanalysis; (d) MODIS; (e) ICAP multi-model ensemble; and (f) VIIRS observation. All panels show AOD at 550 nm.

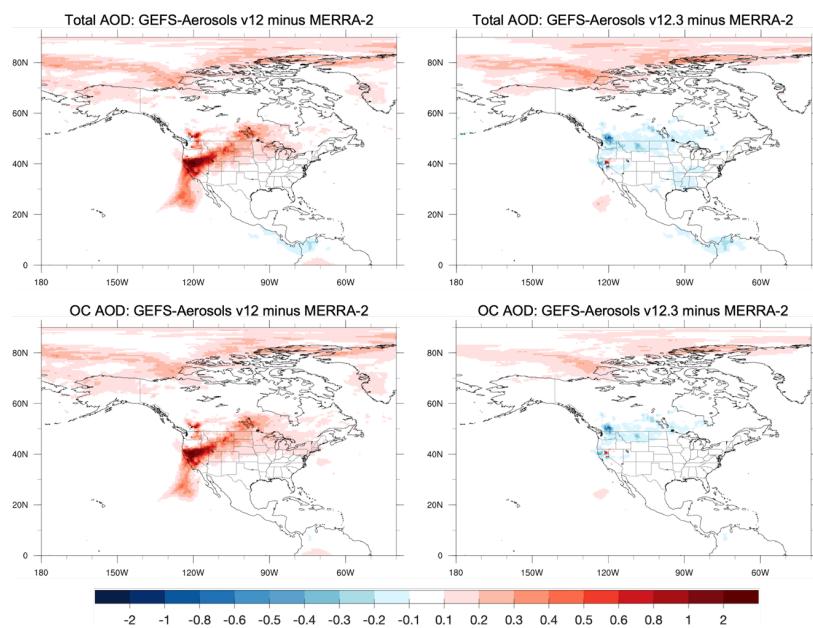
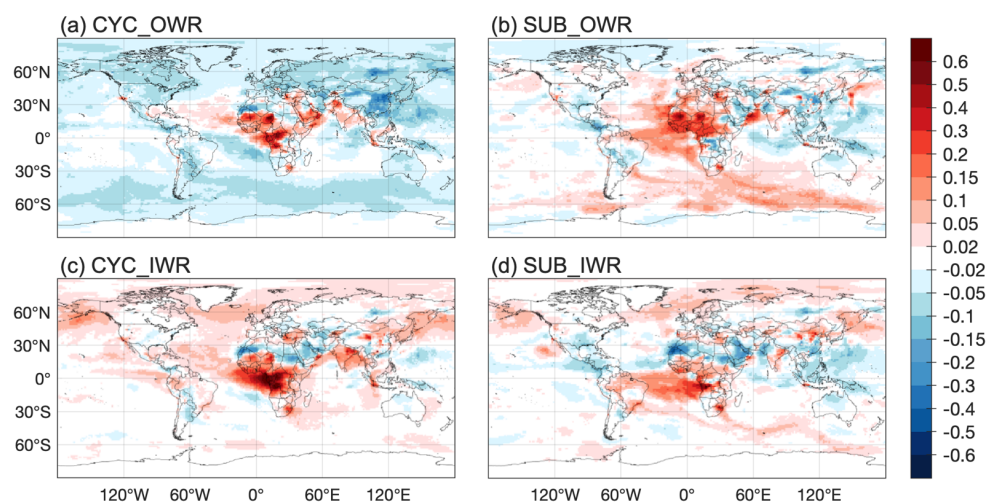


Figure 3. Monthly mean Day-1 Total AOD (top) and OC AOD (bottom) forecast biases for August 2021, defined as GEFS-Aerosols minus MERRA-2, for GEFS-Aerosols v12 (left) and v12.3 (right).

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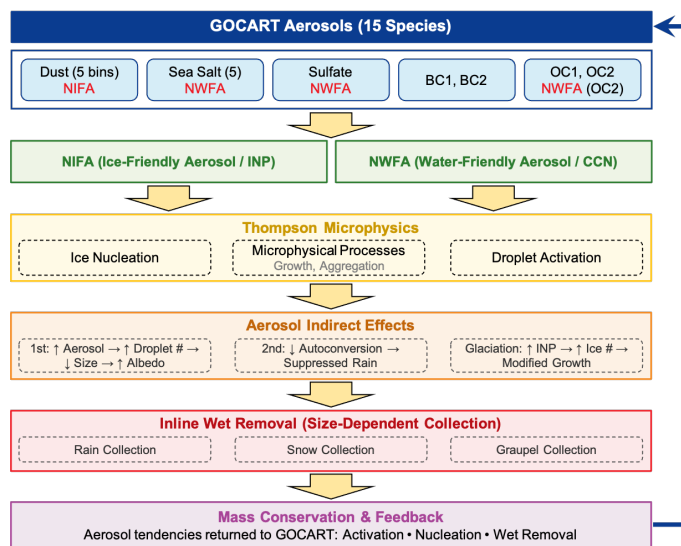


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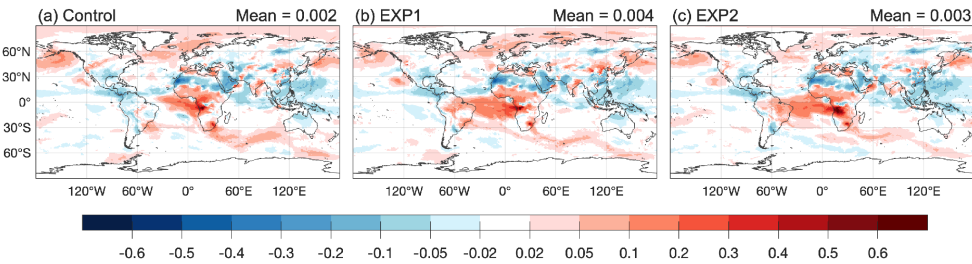


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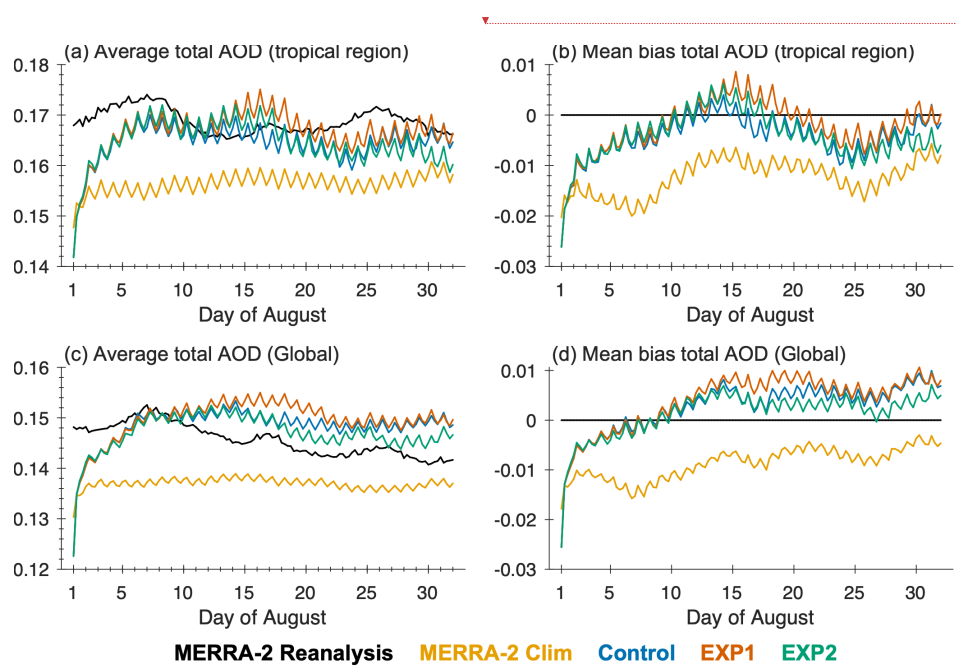
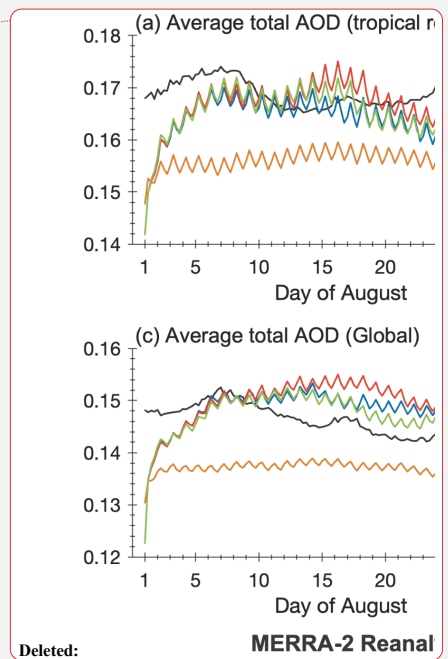
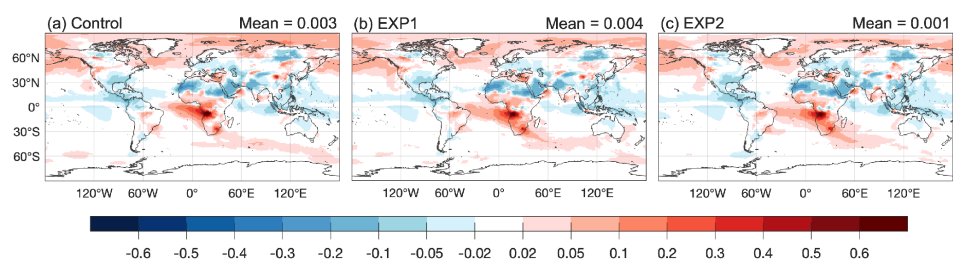


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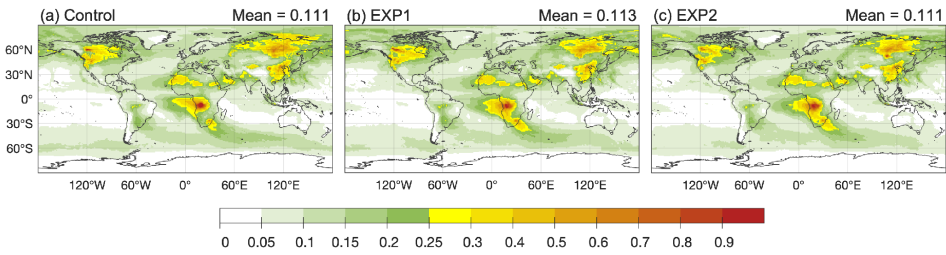
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Figure 8. The same as in Figure 6, but averaged over the six-year of August (2012–2017).

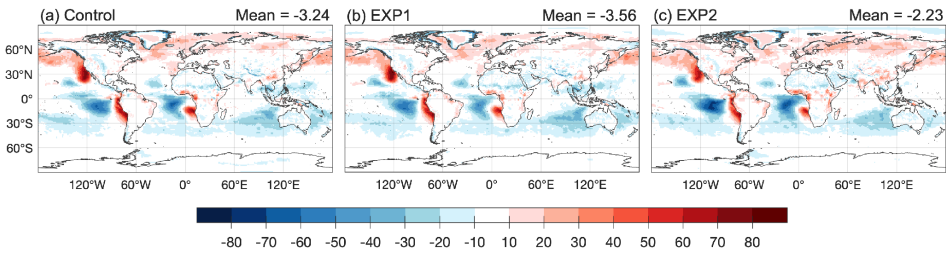
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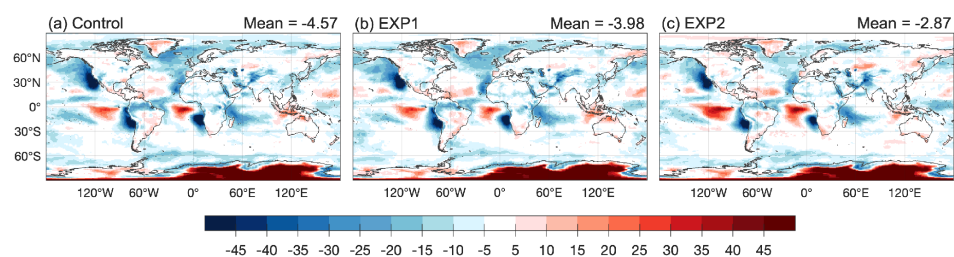
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Figure 10. Six-year averaged (2012–2017) TOA net radiative flux bias compared with CERES for August in UFS-Chem for the (a) Control, (b) EXP1, and (c) EXP2 experiments.

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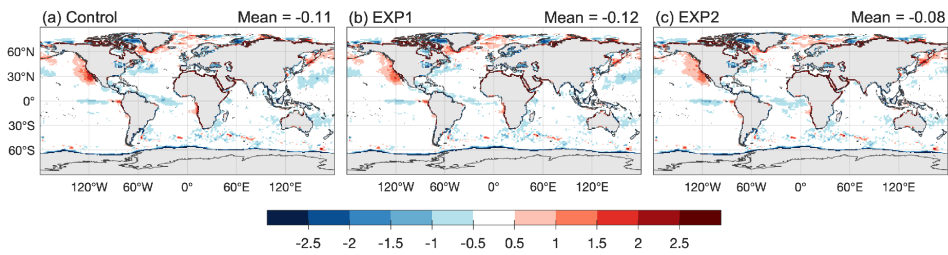


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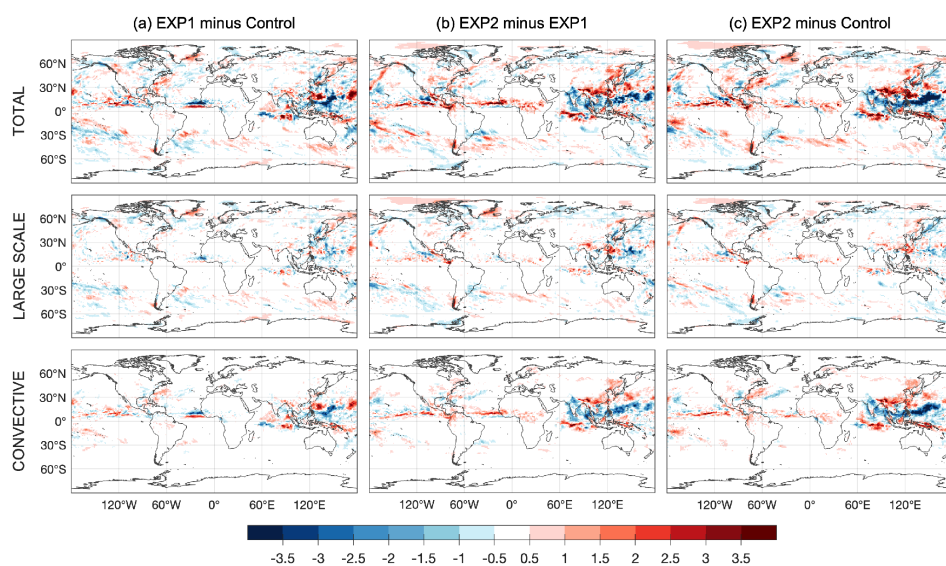


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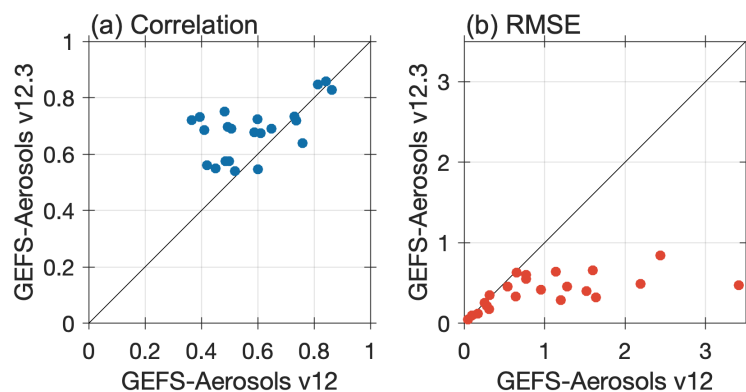


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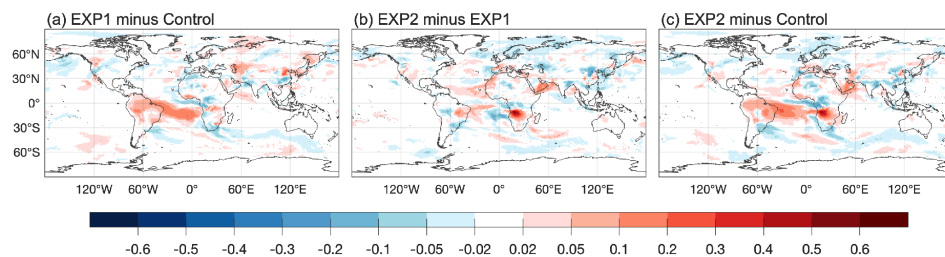


Figure A2. Differences in Total AOD for August 2016 in UFS-Chem. (a) Effect of aerosol radiative feedback (EXP1 minus Control). (b) Aerosol indirect effects (EXP2 minus EXP1). (c) Combined effect of aerosol radiative feedback and aerosol indirect effects (EXP2 minus Control).

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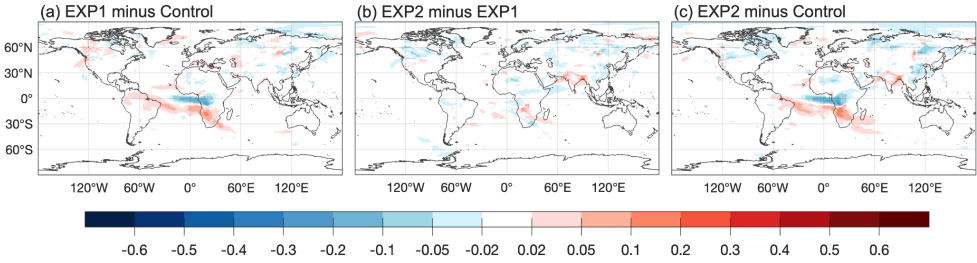


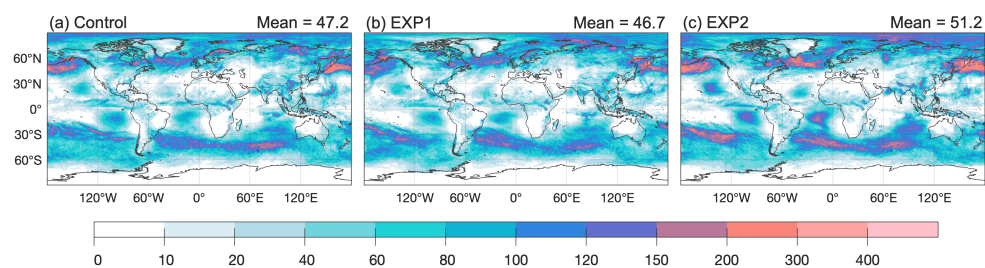
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473 **Table A1.** List of acronyms, abbreviations, and experiment names used in this study.

Acronym / Term	Definition
AERONET	Aerosol Robotic Network
AM4.1	GFDL Atmospheric Model version 4.1
AOD	Aerosol Optical Depth
BC	Black Carbon
CAMS	Copernicus Atmosphere Monitoring Service
CATChem	Configurable ATmospheric Chemistry
CCPP	Common Community Physics Package
CEDS	Community Emissions Data System
CERES	Clouds and the Earth's Radiant Energy System
CMAQ	Community Multiscale Air Quality model
CICE6	Los Alamos Sea Ice Model version 6
CIMEL	CIMEL Sun–Sky Spectral Radiometer
CYC_IWR	24 h cycling experiment with inline large-scale wet removal
CYC_OWR	24 h cycling experiment with offline large-scale wet removal
DAAC	Distributed Active Archive Center
DMS	Dimethyl Sulfide
Dark Target (DT)	Dark Target aerosol retrieval algorithm
Deep Blue (DB)	Deep Blue aerosol retrieval algorithm
EBAF	Energy Balanced and Filled
ECMWF	European Centre for Medium-Range Weather Forecasts
FV3	Finite-Volume Cubed-Sphere dynamical core
GBBEPx	Global Biomass Burning Emissions Product extended (GBBEPx)
GEFS	Global Ensemble Forecast System
GEFS-Aerosols	Global Ensemble Forecast System Aerosols
GEOS	Goddard Earth Observing System
GEOS-CF	Goddard Earth Observing System Composition Forecast
GEOS-FP	Goddard Earth Observing System Forward Processing
GFDL	Geophysical Fluid Dynamics Laboratory
GFS	Global Forecast System
GOCART	Goddard Chemistry Aerosol Radiation and Transport
ICAP-MME	International Cooperative for Aerosol Prediction Multi-Model Ensemble
JMA-MASINGAR	Japan Meteorological Agency Model of Aerosol Species in the Global Atmosphere
LAADS	Level-1 and Atmosphere Archive and Distribution System
MERRA-2	Modern-Era Retrospective analysis for Research and Applications, Version 2
MODIS	Moderate Resolution Imaging Spectroradiometer
MOM6	GFDL Modular Ocean Model version 6
NAAPS	Navy Aerosol Analysis and Prediction System
NASA	National Aeronautics and Space Administration
NASA/LARC/SD/ASDC	NASA Langley Research Center Science Directorate Atmospheric Science Data Center
NCEP	National Centers for Environmental Prediction
NESDIS	National Environmental Satellite, Data, and Information Service
NGACv2	NEMS Global Forecast System Aerosol Component version 2

Acronym / Term	Definition
NMMB/BSC-CTM	Nonhydrostatic Multiscale Model on the B Grid / Barcelona Supercomputing Center Chemical Transport Model
NOAA	National Oceanic and Atmospheric Administration
NRL	Naval Research Laboratory
NUOPC	National Unified Operational Prediction Capability
OC	Organic Carbon
OC2	Hydrophilic Organic Carbon
OISST	Optimum Interpolation Sea Surface Temperature
PBL	Planetary Boundary Layer
PM _{2.5}	Particulate Matter with aerodynamic diameter $\leq 2.5 \mu\text{m}$
RRFS-SD	Rapid Refresh Forecast System – Smoke and Dust
SAS	Simplified Arakawa–Schubert convection scheme
SST	Sea Surface Temperature
SUB_IWR	Fully coupled subseasonal experiment with inline large-scale wet removal
SUB_OWR	Fully coupled subseasonal experiment with offline large-scale wet removal
UFS	Unified Forecast System
TOA	Top of Atmosphere
UFS-Chem	Unified Forecast System coupled with Chemistry
UKMO-UM	United Kingdom Met Office Unified Model
VIIRS	Visible Infrared Imaging Radiometer Suite
WRF-Chem	Weather Research and Forecasting model coupled with Chemistry

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