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3 **Inline Coupling of Simple and Complex Chemistry**
4 **Modules within the Global Weather Forecast model FIM**
5 **(FIM-Chem v1)**
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7 Li Zhang^{1,2 *}, Georg A. Grell², Stuart A. McKeen^{1,3}, Ravan Ahmadov^{1,2}, Karl D. Froyd^{1,3},
8 Daniel Murphy³
9

10 ¹*Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder,*
11 *CO, USA*

12 ²*NOAA/Global Systems Laboratory ([GSL](#)), [Earth System Research Laboratory](#), Boulder, CO, USA*

13 ³*NOAA/Chemical Sciences Laboratory ([CSL](#)), [Earth System Research Laboratory](#), Boulder, CO, USA*
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23 *Correspondence to: Li Zhang (kate.zhang@noaa.gov)

24 CIRES, University of Colorado Boulder

25 GSL EPAD, NOAA ESRL

26 325 Broadway David Skaggs Research Center R/GSL1

27 Boulder, CO 80305

28 1-303-497-3956
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1 **Abstract.**

2 The global Flow-following finite-volume Icosahedral Model (FIM), which was developed in the Global
3 Systems Laboratory (GSL) of NOAA, has been coupled inline with aerosol and gas-phase chemistry schemes
4 of different complexity using the chemistry and aerosol packages from WRF-Chem v3.7, named as FIM-
5 Chem v1. The three chemistry schemes include 1) the simple aerosol modules from the Goddard Chemistry
6 Aerosol Radiation and Transport model that includes only simplified sulfur chemistry, black carbon (BC),
7 organic carbon (OC), and sectional dust and sea salt modules (GOCART); 2) the photochemical gas-phase
8 mechanism RACM coupled to GOCART to determine the impact of more realistic gas-phase chemistry on
9 the GOCART aerosols simulations (RACM_GOCART); and 3) a further sophistication within the aerosol
10 modules by replacing GOCART with a modal aerosol scheme that includes secondary organic aerosols (SOA)
11 based on the VBS approach (RACM_SOA_VBS). FIM-Chem is able to simulate aerosol, gas-phase chemical
12 species and SOA at various spatial resolutions with different levels of complexity and quantify the impact of
13 aerosol on numerical weather predictions (NWP). We compare the results of RACM_GOCART and
14 GOCART schemes which uses the default climatological model fields for OH, H₂O₂, and NO₃. We find
15 significant reductions of sulfate that are on the order of 40% to 80% over the eastern US and are up to 40%
16 near the Beijing region over China when using the RACM_GOCART scheme. We also evaluate the model
17 performance by comparing with the Atmospheric Tomography Mission (ATom-1) aircraft measurements in
18 2016 summer. FIM-Chem shows good performance in capturing the aerosol and gas-phase tracers. The model
19 predicted vertical profiles of biomass burning plumes and dust plumes off the western Africa are also
20 reproduced reasonably well.

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1 **1 Introduction**

2 The impacts of aerosol on weather and climate are generally attributed to the direct, semidirect, indirect, and
3 surface albedo effects, with the direct effect predominating radiative forcing over a global scale [e.g. *Bauer*
4 *and Menon, 2012*]. However, there are significant differences in estimates of direct aerosol radiative forcing
5 between various global aerosol models, particularly with respect to the attribution of forcing to specific
6 aerosol species and sources [*Myhre et al., 2013*]. Discrepancies in direct radiative forcing are also found
7 between global aerosol model results and determinations based on satellite retrievals, with assumptions
8 related to aerosol composition and optical properties as the primary source of difference [e.g. *Su et al., 2013*].
9 Several processes and steps are necessary to accurately include aerosol effects within a meteorological
10 forecast. Aerosol abundance, composition, and size distribution are the basic quantities needed within
11 calculations of the optical properties, which in turn are used within radiative transfer calculations to calculate
12 heating or cooling rates and are incorporated within the thermodynamic calculations of the numerical forecast.
13 The importance of aerosol impacts on the meteorological fields for climate modeling have been widely
14 recognized by many studies [e.g. *Xie et al., 2013*; *Yang et al., 2014*; *Wang et al., 2014a, 2014b*; *Colarco et*
15 *al., 2014*]. Since it is increasingly common for modeling systems to start using prognostic online aerosol
16 schemes and more accurate emissions, many studies exist that show the importance of including aerosols at
17 least for case studies or over limited time periods. On NWP timescales (5–10 days), *Rodwell and Jung [2008]*
18 showed an improvement in forecast skill and general circulation patterns in the tropics and extra-tropics by
19 using a monthly varying aerosol climatology rather than a fixed climatology in the European Centre for
20 Medium-Range Weather Forecasting (ECMWF) global forecasting system. The inclusion of the direct and
21 indirect effects of aerosol complexity into a version of the global NWP configuration of the Met Office
22 Unified Model (Met UM) shows that the prognostic aerosol schemes are better able to predict the temporal
23 and spatial variations of atmospheric aerosol optical depth, which is particularly important in cases of large
24 sporadic aerosol events such as large dust storms or forest fires [*Mulcahy et al., 2014*]. The aerosols from
25 biomass burning sources have been shown to have an effect on large-scale weather patterns within global
26 scale models [e.g. *Sakaeda, 2011*] and synoptic scale meteorology within the WRF-Chem regional model
27 [*Grell et al., 2011*]. *Toll et al. [2015]* showed considerable improvement in forecasts of near-surface
28 conditions during Russian wildfires in summer 2010 by including the direct radiative effect of realistic
29 aerosol distributions. Likewise, many global models [e.g. *Haustein et al., 2012*] and regional models [e.g.
30 WRF-Chem, *Zhao et al., 2010*] have established a clear connection between dust emissions and weather
31 patterns over synoptic to seasonal time scales. While positive impacts of predicted aerosols on weather
32 forecasts have been shown on an episodic basis, a systematic verification of current state-of-the-art
33 operational modeling systems does not yet demonstrate that the impact is statistically significant over longer
34 periods of time to warrant the required additional computational resources [*Peuch et al., 2014*]. Operational
35 forecast systems are usually highly tuned and still use aerosol climatologies. The inclusion of aerosols in the
36 presence of strong sources or sinks should lead to an improvement of predictive skills. A successful example
37 of a short-range weather forecasting coupled with the smoke tracer is the High-Resolution Rapid Refresh

1 coupled with Smoke (HRRR-Smoke) model [Ahmadov et al, 2017]. The model forecasts 3D smoke
2 concentrations and its radiative impacts over the CONUS domain at 3km spatial gridding
3 [<https://rapidrefresh.noaa.gov/hrrr/HRRRsmoke/>].
4 By applying the chemistry package from WRF-Chem v3.7 into the Flow-following finite-volume Icosahedra
5 Model (FIM, Bleck et al. 2015), named as FIM-Chem v1, we essentially make it possible to explore the
6 importance of different levels of complexity in gas and aerosol chemistry, as well as in physics
7 parameterizations on the interaction processes in global modeling systems. FIM is used in the sub-seasonal
8 experiment (SUBx) for sub-seasonal to seasonal (S2S) forecasting and is now considered a steppingstone
9 towards NOAA's Next Generation Global Prediction System, which will be based on the third generation
10 non-hydrostatic Finite Volume Cubed Sphere (FV3) dynamic core [Sun et al., 2018a, b]. The chemistry
11 component created here is designed to be moved flawlessly into FV3. WRF-Chem currently has 63 different
12 gas and aerosol chemistry options, as well as several microphysics and radiation parameterizations, which
13 are coupled to chemistry to simulate direct and indirect aerosol feedback processes. In this study we
14 demonstrate three examples of different complexities on the aerosol forecasts by FIM-Chem. The current
15 real-time forecast uses simple bulk aerosol modules from the GOCART model, with a simplified chemistry
16 for sulfate production. This chemistry scheme does not include NOx/Volatile Organic Compounds (VOC)
17 gas chemistry or SOA formation. Currently the real-time GOCART application uses climatological fields of
18 OH, H₂O₂ and NO₃ to drive the oxidation of SO₂ and oceanic dimethyl sulfide to sulfate.
19 Here we also investigate the sensitivity to the addition of complex gas-phase chemistry and a more reasonable
20 inclusion of Secondary Organic Aerosol formation. Organic matter makes up the significant fraction of the
21 sub-micron aerosol composition [Zhang et al., 2007], and organic aerosol (OA) along with sulfate and black
22 carbon are believed to be the main anthropogenic contributors to direct radiative forcing on a global scale
23 [Myhre et al., 2013]. A computationally efficient SOA parameterization based on the Volatility Basis Set
24 approach [Donahue, 2011] was implemented in WRF-Chem by Ahmadov et al. (2012).
25 To evaluate the model performance, the observation data from the NASA Atmospheric Tomography aircraft
26 mission (ATom-1, 2016) is used, in which the DC-8 is instrumented to make high-frequency in situ
27 measurements of the most the chemical species over the Pacific and Atlantic Oceans, and across the Arctic
28 and US, to evaluate the model performance. Section 2 describes some aspects of the FIM and FIM-Chem
29 model, the coupling of aerosol configurations, gas-phase chemical schemes and an overview of the
30 observation data used to evaluate the model results. The chemical weather forecasts by using three different
31 gas and aerosol chemistry schemes with different level of complexities are shown in Section 3. Section 4
32 presents the evaluations of the chemical weather forecasts, and the model evaluations are investigated in
33 Section 5. We end with discussion and conclusions in Section 6.

34 **2 Models and Observation**

35 **2.1 FIM**

1 FIM is a hydrostatic global weather prediction model based on an icosahedral horizontal grid and a hybrid
2 terrain following/isentropic vertical coordinate [Bleck et al., 2015]. Icosahedral grids are generated by
3 projecting an icosahedron onto its enclosing sphere and iteratively subdividing the 20 resulting spherical
4 triangles until a desired spatial resolution is reached. The main attraction of geodesic grids lies in their fairly
5 uniform spatial resolution and in the absence of the two pole singularities found in spherical coordinates. The
6 primary purpose of using a near-isentropic vertical coordinate in a circulation model is to assure that
7 momentum and mass field constituents (potential temperature, moisture, chemical compounds, etc.) are
8 dispersed in the model in a manner emulating reality, namely, along neutrally buoyant surfaces. The FIM
9 model has been tested extensively on real-time medium-range forecasts to ready it for possible inclusion in
10 operational multi-model ensembles for medium-range to seasonal prediction, and the following simulations
11 are performed at G6 (~128 km) horizontal resolution.

12 In FIM-Chem, the column physics parameterizations have been taken directly from the 2011 version of the
13 GFS [Bleck et al., 2015]. The physical parameterizations include the Grell-Freitas convection
14 parameterization [Grell and Freitas, 2014], the Lin et al. [1983] cloud microphysics scheme, coupled to the
15 model aerosol parameterization and modified to include second moment effects, and the land surface
16 processes simulated by the NCEP's Noah land surface model [Koren et al. 1999 and Ek et al. 2003].

17 **2.2 FIM-Chem**

18 FIM-Chem, is a version of the FIM model coupled inline with chemical transport model including three
19 aerosol and gas-phase chemistry schemes of different complexities, where physics and chemistry components
20 of the model are simulated simultaneously. The chemical modules and coupling schemes are adopted from
21 the WRF-Chem model v3.6.1 [Grell et al. 2005; Fast et al. 2006; Powers et al., 2017]. [The different three](#)
22 [chemical schemes have been listed in Table 1 for comparisons.](#)

23 **2.2.1 GOCART scheme**

24 The first chemical option is the simplest aerosol modules that from the GOCART model, which includes
25 simplified sulfur chemistry for sulfate simulation from chemical reactions of SO₂, H₂O₂, OH, NO₃ and DMS,
26 bulk aerosols of black carbon (BC), organic carbon (OC), and sectional dust and sea salt. For OC and BC,
27 hydrophobic and hydrophilic components are considered and the chemical reactions using prescribed OH,
28 H₂O₂, and NO₃ fields for gaseous sulfur oxidations [Chin et al., 2000]. The dust scheme is using the Air
29 Force Weather Agency (AFWA) scheme with five dust size bins [LeGrand et al., 2019]. The bulk vertical
30 dust flux is based on the Marticorena and Bergametti scheme [Marticorena et al., 1995], whereas the particle
31 size distribution is built according to Kok, 2011, which is based on the brittle material fragmentation theory.
32 Four size bins are considered for the sea salt simulation. The sea salt emissions from the ocean are highly
33 dependent on the surface wind speed [Chin et al., 2000]. [There are totally 19 chemical tracers for transport](#)
34 [and 4 chemical reactions in the GOCART schemes. For 24 hours forecast, it takes about 4 minutes.](#)

1 2.2.2 RACM_GOCART scheme

2 The simple GOCART aerosol scheme does not include photolysis, full gas chemistry and secondary organic
3 aerosol production, and it normally uses climatological fields of OH, H₂O₂ and NO₃ to drive the oxidation of
4 SO₂ and oceanic dimethyl sulfide (DMS) to produce sulfate. Based on the GOCART aerosol module, the
5 second chemical option includes the photochemical gas-phase mechanism of Regional Atmospheric
6 Chemistry Mechanism (RACM), which is able to determine the impact of the additional gas-phase
7 complexity on the aerosol simulations (RACM_GOCART). The RACM chemistry mechanism is based upon
8 the earlier Regional Acid Deposition Model, version 2 (RADM2) mechanism [Stockwell et al., 1990] and the
9 more detailed Euro-RADM mechanism [Stockwell and Kley, 1994]. It includes a full range of photolysis,
10 biogenic VOCs, full NO_x/VOC chemistry, inorganic and organic gaseous species to perform air pollution
11 studies that includes rate constants and product yields from the laboratory measurements [Stockwell et al.,
12 1997]. The simplified sulfur chemistry for sulfate formation does not use climatological fields of OH, H₂O₂
13 and NO₃ from GOCART model to drive the oxidation of SO₂ as that in GOCART, and it is replaced by
14 explicitly simulating the gas-phase RACM chemistry. Meanwhile, the SO₂ is also impacted by the RACM
15 gas-phase chemistry, leading to differences with the GOCART simulations. There are 214 chemical reactions
16 and 68 chemical tracers for transport in the RACM_GOCART scheme. It takes about 19 minutes for a 24
17 hours forecast.

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18 2.2.3 RACM_SOA_VBS scheme

19 Other than the simple GOCART aerosol scheme in both GOCART and RACM_GOCART, we implemented
20 a more complex gas-aerosol chemistry scheme of RACM_SOA_VBS in FIM-Chem. This scheme includes
21 the RACM based gas chemistry and the modal aerosol scheme, MADE (Modal Aerosol Dynamics Model
22 for Europe) with SOA based on the VBS (Volatility Basis Set) approach [Ahmadov et al., 2012]. The
23 RACM_SOA_VBS scheme includes photolysis reactions for multiple species, full nitrogen and VOC
24 (anthropogenic and biogenic) chemistry, inorganic and organic aerosols. All the secondary gas species
25 required for the SO₂ oxidation are simulated explicitly by the gas chemistry scheme here. There are 233
26 chemical reactions and 103 transported chemical tracers in the RACM_SOA_VBS scheme. It takes about 22
27 minuets for 24 hours forecast. The new SOA mechanism contains four volatility bins for each SOA class,
28 and their organic vapors that condense onto aerosol. Equilibrium between gas and particle phase matter for
29 each bin is assumed in the model. The SOA species are added within the MADE aerosol module, which
30 considers composition within the Aitken and the accumulation modes separately. The VBS approach was
31 included for SOA production, updated SOA yields, and multigenerational VOC oxidation. The VOCs
32 forming SOA are divided into two groups, anthropogenic and biogenic. Isoprene, monoterpenes and
33 sesquiterpenes are emitted by biogenic sources, while other VOCs by anthropogenic sources. More detailed
34 descriptions about the VBS approach based on SOA scheme can be found in Ahmadov et al., 2012.

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35 2.2.4 Emission, deposition, and aerosol optical properties

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1 The preprocessor PREP-CHEM-SRC v1.5 [Freitas et al., 2011], a comprehensive tool aiming at preparing
2 emission fields of the chemical species for use in atmospheric-chemistry transport models, is used
3 to generate the emissions for FIM-Chem. It includes the Hemispheric Transport of Air Pollution
4 (HTAP) v2 global anthropogenic emission inventory [Janssens-Maenhout et al., 2015] and biogenic VOC
5 emissions simulated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.0
6 parameterization [Guenther et al., 2006]. The diurnal variability based on a function of anthropogenic
7 activities is applied to the HTAP emissions and the diurnal cycle of solar radiation and air temperature is
8 applied to the biogenic emissions. The biomass burning emission estimated by the Brazilian Biomass Burning
9 Emissions Model [3BEM, Longo et al. 2010; Grell et al., 2011] is also included in the PREP-CHEM-SRC.
10 The 3BEM is based on near real-time remote sensing fire products to determine fire emissions and plume
11 rise characteristics [Freitas et al., 2007; Longo et al., 2010]. Although the same settings are used for these 3
12 schemes in PREP-Chem-SRC, the speciation profiles are modified for each specific mechanism. The fire
13 emissions are updated as they become available and are spatially and temporally distributed according to the
14 fire count locations obtained by remote sensing of Moderate Resolution Imaging Spectroradiometer
15 (MODIS) onboard Terra and Aqua satellites [Giglio et al., 2003]. The biomass burning emission factors are
16 from Andreae and Merlet [2001]. Over the CONUS domain the MODIS data are replaced by the Wildfire
17 Automated Biomass Algorithm (WF_ABBA) processing system. The WF_ABBA is able to detect and
18 characterize fires in near real-time, providing users with high temporal and spatial resolution fire detection
19 data (<http://www.ssd.noaa.gov/PS/FIRE/Layers/ABBA/abba.html>). In the current retrospective forecast of
20 2016, there is no day lag input for emission in the model. A one-dimension (1-D) time-dependent cloud
21 model implemented to calculate injection heights and emission rates online in all of the three chemical
22 schemes [Freitas et al., 2007].
23 Similar to WRF-Chem model, the flux of gases and aerosols from the atmosphere to the surface is calculated
24 by multiplying concentrations of the chemical species in the lowest model layer by the spatially and
25 temporally varying deposition velocities, the inverse of which is proportional to the sum of three
26 characteristic resistances (aerodynamic resistance, sublayer resistance, surface resistance [Grell et al. 2005].
27 The GOCART aerosol dry deposition includes sedimentation (gravitational settling) as a function of particle
28 size and air viscosity and surface deposition as a function of surface type and meteorological conditions
29 [Wesely, 1989]. The dry deposition of sulfate is described differently. In the case of simulations without
30 calculating aerosols explicitly, sulfate is assumed to be presented in the form of aerosol particles, and the dry
31 deposition of aerosol and gas phase species is parameterized as described in Erisman et al. [1994]. For
32 RACM_SOA_VBS chemical option, the dry deposition velocity of the organic condensable vapors (OCVs)
33 is parameterized as proportional to the model calculated deposition velocity of a very soluble gas, nitric acid
34 (HNO₃). The parameter which determines the fraction (denoted as “depo_fact”) of HNO₃ is assumed in the
35 model since no observation constraints are available. The dry deposition velocity of HNO₃ is calculated by
36 the model during runtime [Ahmadov et al., 2012]. Wet deposition accounts for the scavenging of aerosols in

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1 convective updrafts and rainout/washout in large-scale precipitation [Giorgi and Chameides, 1986; Balkanski
2 et al., 1993].

3 The aerosol optical properties such as extinction, single-scattering albedo, and the asymmetry factor for
4 scattering are computed as a function of wavelength. Each chemical constituent of the aerosol is associated
5 with a complex index of refraction. A detailed description of the computation of aerosol optical properties
6 can be found in Fast et al. [2006] and Barnard et al. [2010].

7 2.3 Observations

8 The Atmospheric Tomography Mission (ATom) studies the impact of human-produced air pollution on
9 greenhouse gases and on chemically reactive gases in the atmosphere [Wofsy et al., 2018]. ATom deploys
10 instrumentation to sample the atmospheric composition, profiling the atmosphere in 0.2 to 12 km altitude
11 range. Flights took place in each of 4 seasons over a 22-month period. They originated from the Armstrong
12 Flight Research Center in Palmdale, California, flew north to the western Arctic, south to the South Pacific,
13 east to the Atlantic, north to Greenland, and returned to California across central North America over the
14 Pacific and Atlantic oceans from ~ 80°N to ~ 65°S. ATom establishes a single, contiguous global-scale data
15 set. This comprehensive data set is used to improve the representation of chemically reactive gases and short-
16 lived climate forcers in global models of atmospheric chemistry and climate. Comparisons of model forecasts
17 with 5 flights from the first ATom mission (August 15–23, 2016) are shown here as examples of model
18 performance for specific events, such as wildfires and dust-storms, or specific conditions such as oceanic
19 versus continental.

20 The Particle Analysis by Laser Mass Spectrometry (PALMS) instrument samples the composition of single
21 particles in the atmosphere with diameters within ~150 nm - 5 μm range. It measures nearly all components
22 of aerosols from volatiles to refractory elements, including sulfates, nitrates, carbonaceous material, sea salt,
23 and mineral dust [Murphy et al., 2006]. The PALMS instrument was originally constructed for high-altitude
24 sampling [Thomson et al., 2000; Murphy et al., 2014] and has since been improved and converted for other
25 research aircraft. Uncertainty in mass concentration products is driven mainly by particle sampling statistics.
26 Relative 1-sigma statistical errors of 10–40% are typical for each 3-min sample at a mass loading of 0.1 ug/m3

27 [Froyd et al., 2019]. In August 2016, PALMS was sampling on the NASA DC-8 aircraft as part of the ATom
28 program (<https://espo.nasa.gov/missions/atom/content/ATom>). Aerosol composition determinations using
29 the PALMS instrument during ATom have been described and interpreted previously [Murphy et al., 2018,
30 2019; Schill et al., 2020; Bourgeois et al., 2020]. The PALMS mass concentrations for various species are
31 derived by normalizing the fractions of particles of each size and type to size distributions measured by
32 optical particle counters [Froyd et al., 2019].

33 Figure 1 shows the vertical profiles and transect time series of the ATom-1 flight tracks on August 15th and
34 17th, 2016 over Atlantic Ocean on August 23rd, 2006 over US. The August 15th flight originates from the
35 southwestern Atlantic and ends near the southern equatorial Atlantic; the August 17th flight is from the
36 southern equatorial Atlantic to the northern Atlantic; and the August 23rd flight is from Minnesota to Southern

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1 California. For analysis and model validations, here we mark 16 vertical tracks and 3 horizontal tracks for
2 August 15th, 16 vertical tracks and 2 horizontal tracks for August 17th, and 8 vertical tracks and 4 horizontal
3 tracks for August 23rd.

4 3 Chemical Composition Forecast

5 ~~We perform a 5-days forecast started from 00:00 UTC July 29th 2016, and get the predicted results at 00:00~~
6 ~~UTC August 3rd 2016 in Fig.2 and Fig.3.~~ For the aerosol forecast, the GOCART and RACM_GOCART
7 scheme are quite similar since they are using the same GOCART aerosol module. However, the major
8 difference is the impact of including gas-phase chemistry on aerosol. The simpler GOCART package uses
9 climatological fields for OH, H₂O₂, and NO₃ from previous GEOS model simulations, while these species
10 are explicitly simulated in the RACM_GOCART chemistry mechanism. The PM_{2.5} concentrations are the
11 sum of BC, OC, sulfate, the fine bins (diameter < 2.5 micrometers) of dust and sea salt. The forecast aerosol
12 results of surface PM_{2.5} and sulfate using GOGART and RACM_GOCART and their differences
13 (RACM_GOCART minus GOCART) are showed at Fig. 2. The general patterns of surface PM_{2.5} are quite
14 similar in these two schemes, with the maximum surface concentrations of more than 100 µg/m³ over the
15 dust source region of western Africa, ~~part of the southern African fire regions and part of the~~ polluted areas
16 of south Asia and eastern China. However, the surface concentrations of PM_{2.5} in GOCART and
17 RACM_GOCART (the latter minus former) show substantial differences, decreasing more than 15 µg/m³
18 over eastern US and 20 µg/m³ over eastern China, when using the RACM_GOCART scheme. The main
19 factor that contributes to the significant differences of surface PM_{2.5} concentration is sulfate (~~see Fig.2 right~~
20 ~~column~~). The maximum surface sulfate concentrations are over the eastern US, India and eastern China. We
21 find the reductions of sulfate are about 10 µg/m³ on the order of 40-80% over the eastern US and are up to
22 40% over eastern China in RACM_GOCART (Fig. 2b). The major differences for sulfate production ~~between~~
23 ~~GOCART and GOCART-RACM are the background fields of H₂O₂, OH and NO₃, GOCART uses the model~~
24 ~~climatological backgrounds fields of H₂O₂, OH and NO₃ while GOCART-RACM uses the online calculated~~
25 ~~fields of H₂O₂, OH and NO₃ from the RACM mechanism.~~

26 Fig. 3 shows the comparisons of surface H₂O₂, OH, and NO₃ between GOCART and RACM_GOCART
27 schemes. Globally the prescribed surface H₂O₂ in GOCART is generally larger than that explicitly simulated
28 by RACM_GOCART. The maximum of surface H₂O₂ over Africa, India and eastern Asia show
29 significant diversity. The explicitly real-simulated instantaneous surface H₂O₂ in RACM_GOCART is much
30 lower, by 40-60% over India and eastern Asia and 20% over eastern US, while much higher (> 80%) over
31 middle Africa, northeastern regions of Canada, and northwestern areas of South America. Even though the
32 patterns of surface OH are quite comparable in the GOCART and RACM_GOCART schemes ~~at 00 UTC~~,
33 the real-simulated instantaneous surface OH is 80% lower over eastern China when using the
34 RACM_GOCART scheme. The other big difference is over the western US with the simulated surface OH
35 in RACM_GOCART being much higher over northwestern US and lower over the southwestern US ~~at 00~~

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1 UCT. The surface NO₃ differences are mainly over the Africa and north Indian Ocean, that the real-simulated
2 instantaneous surface NO₃ is much larger using the RACM_GOCART scheme at 00 UCT. Since surface
3 H₂O₂ and OH are the major species converting SO₂ to sulfate, their decreases cause sulfate reductions over
4 broad areas. The OH differences of GOCART and RACM_GOCART schemes at 12 UTC shows reduction
5 over Africa, India and Asia, corresponding to the decreasing sulfate over those aeras, accounting for the
6 major differences in sulfate production between the two mechanisms.

7 The RACM_GOCART model is able to predict gas phase species by using the RACM gas-phase mechanism.
8 Ozone (O₃) and other gas pollutants are determined by the emissions of nitrogen oxides and reactive organic
9 species, gas- and aqueous-phase chemical reaction rates, depositions, and meteorological conditions. Fig. 4
10 represents the 5-days surface O₃ forecast globally at 12:00 UTC August 2nd and 00:00 UTC August 3rd, 2016,
11 which started from 00:00 UTC July 29th, 2016. Similar to other studies, a lot of chemical transport models
12 (CTMs) tend to significantly overestimate surface O₃ in the southeast US [Lin et al., 2008; Fiore et al., 2009;
13 Reidmiller et al., 2009; Brown-Steiner et al., 2015; Canty et al., 2015; Travis et al., 2016], which is an
14 important issue for the design of pollution control strategies [McDonald-Buller et al., 2011]. We see similar
15 problem in FIM-Chem that the predicted surface O₃ concentration on 00:00 UTC August 3rd, 2016 is also
16 overestimated (see Fig. 4b). The relative low surface O3 is likely due to the O3 titration during the early
17 morning and nighttime periods. It well known that the O₃ production involves complex chemistry driven by
18 emissions of anthropogenic nitrogen oxide radicals (NO_x=NO+NO₂) and isoprene from biogenic emissions.
19 The primary basis of O₃ may be due to the inventory of HTAP v2 anthropogenic emission over North America,
20 which is from U.S. EPA's 2005 National Emission Inventory (NEI2005). A few studies have pointed out that
21 the NO_x emissions in the NEI-2005 and NEI-2011 from the EPA is too high [Brioude, 2011; Travis et al.,
22 2016] over the US. It must be reduced by 30-60% from mobile and industrial sources in the NEI 2011
23 inventory [Katherine et al., 2016], while the NO_x emissions over United States should be reduced more for
24 2016 simulation since the NEI2005 NO_x emission is much larger than that of NEI2011
25 (<https://cfpub.epa.gov/roe/>). Also, the dry depositions of ozone, isoprene emissions and in the loss of NO_x
26 from formation of isoprene nitrates could also result into these overestimations [Lin et al., 2008; Fiore et al.,
27 2005].

28 The SOA parameterization based on the volatility basis and VBS approach applied within FIM-Chem, has
29 the ability to simulate and predict SOA using the RACM_SOA_VBS scheme [Ahmadov et al., 2012], which
30 include the anthropogenic secondary organic aerosols (ASOA) and biogenic secondary organic aerosols
31 (BSAO) for both the nucleation and accumulation modes. Fig. 5 shows the predicted SOA at 12:00 UTC
32 August 2nd and 00:00 UTC August 3rd, 2016. The maximum surface SOA concentrations are over southern
33 Africa, which may be caused by the wildfire emissions. The Eastern US, western Europe and eastern Asia
34 are the other high SOA concentrations areas. There is not significant diurnal variability for the SOA spatial
35 distributions, and the diurnal cycle of fire emission has not been included.

36 4 Using ATom-1 observations to evaluate the FIM-Chem Model

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1 The retrospective daily forecast uses cycling for the chemical fields since no data assimilation is included in
2 the chemical model. Meteorological fields are initialized by the GFS meteorological fields every 24 hours,
3 while the chemical fields from the last output (forecast at 24:00 UTC) are used as the initial conditions of the
4 current forecast (00:00 UTC). Stratospheric O₃ above tropopause is taken from satellite derived fields
5 available within GFS. For the ATom-1 forecast periods, considering there is no chemical initial conditions,
6 we performed a two-week spin-up period (from July 15th to July 28th) before the first observational
7 comparison day (July 29th, 2016) to help get a realistic chemical initial conditions for the ATom-1 forecast
8 period. It should be noted that stratospheric chemistry is incomplete (no halogen chemistry) in the model.

9 In this section, we compare 24 hours forecasts of FIM-Chem for the major aerosols and gas tracers for the
10 three different chemical schemes listed above. The FIM-Chem model results are sampled at the grid with
11 nearest latitude and longitude, and interpolated logarithmically in altitude according to the ATom-1
12 measurements. Temporally, 1-second measurements are matched to the nearest hour of the FIM-Chem hourly
13 model output, which translates into a spatial uncertainty of ~ 128 km, or ~1 model grid cell, for typical DC-
14 8 airspeeds.

15 **4.1 Comparisons of the gas and aerosols species between FIM-Chem and the ATom-1 measurements** 16 **over Atlantic**

17 The comparison between RACM_GOCART and RACM_SOA_VBS schemes for the chemical species, e.g.,
18 EC (elemental carbon, which is the same as BC), CO and O₃, that are mainly affected by the biomass burning
19 emissions from wildfires during August 15th and August 17th, are shown in Fig. 6. The model shows very
20 good performance in reproducing the profiles of EC and CO, especially capturing the biomass burning
21 plumes near the tropics. But it also shows some differences for EC in the results of GOCART (figures not
22 shown here since it is almost the same as that of RACM_GOCART) and RACM_GOCART schemes above
23 4~5 km, where model results are overestimated. Generally, the EC performance of RACM_GOCART is
24 much better at low altitudes but has a high biased at high altitudes where the RACM_SOA_VBS
25 performs well. After investigating, we noticed that the GOCART and RACM_GOCART aerosol modules
26 both assume there is no wet deposition for externally-mixed, hydrophobic BC, only for hydrophilic BC. This
27 assumption would result into the overestimation of EC at higher levels due to less wash out of hydrophobic
28 BC. Other models with simple wet removal schemes have shown similar overestimation of EC in the upper
29 troposphere (Schwarz et al., 2013; Yu et al., 2019). However, aerosols in the RACM_SOA_VBS scheme are
30 internally mixed, so there is a much larger wet deposition, and less EC in the upper levels. This an important
31 difference about the carbonaceous aerosol for both hydrophobic BC and OC in the wet removal. The
32 comparison with the observations provides a good resource for further improvements within the wet removal
33 parameterization. The second column in Fig. 6 compares CO for the observations, RACM_GOCART and
34 RACM_SOA_VBS schemes. Overall, the forecast is able to capture the observed latitude-height profiles of
35 CO mixing ratio. However, they both show high biases at low altitude (about ~2km) in the tropics. Other
36 than that, there are still some differences such as the underestimated CO mixing ratio above 6 km over the

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1 tropics ~~and overestimate~~ near the surface. ~~Also, the model does not reproduce the fire plume height correctly~~
 2 for the biomass burning ~~emissions~~ over this area, which may be due to ~~vertical transport~~ or lower injection
 3 heights ~~near the fire source region~~. For O₃, the model is able to consistently capture O₃ mixing ratios with
 4 both RACM_GOCART and RACM_SOA_VBS schemes, including the stratospheric intrusion near 40°S at
 5 about 9 km height, though it is slightly higher near 40°N at about 12 km height. We find that over equatorial
 6 areas at about 2-4 km height, the modeled O₃ mixing ratio is underestimated by about 30%. This may also
 7 relate to the injection height of biomass burning that resulted ~~in~~ much lower CO at this altitude, since CO is
 8 ~~an important precursor for O₃ production. Near the surface the overpredicted CO in the RACM_GOCART~~
 9 ~~and RACM_SOA_VBS schemes does not result in high O₃. It may be related to other O₃ precursors other~~
 10 ~~than CO, such as missing VOC and NO_x sources. Large uncertainties in both the biogenic and anthropogenic~~
 11 ~~emission inventories are expected over Western Africa.~~ Besides the aerosol and gas tracers associated with
 12 the biomass burning emissions, we also compare the HCHO, OH and H₂O₂, which are the important
 13 precursors or oxidants to many other species within the RACM_GOCART and RACM_SOA_VBS schemes
 14 (see Fig. 7). Generally, the pattern of the modeled HCHO mixing ratio is almost the same as that of the
 15 ATom-1 measurements. The variations from south to north are captured by these two schemes except a little
 16 underestimation near about 10 km height. For OH, the model reproduces the vertical and temporal variations,
 17 including the large mixing ratios over the northern hemisphere. Some slight differences are apparent, e.g.,
 18 the overestimates over 44°S at 3-9 km height and the underestimates over 40°N above 10 km height. Similarly,
 19 there is more spatial variability in the ATom-1 measurement of H₂O₂. Above 6km the model overestimates
 20 H₂O₂ south of 40°S and overestimates from 20°S to the northern hemisphere above 6 km. Overall, the model
 21 and ATom-1 measurement are more consistent at lower altitudes for H₂O₂.
 22 Figures 8 and 9 show more detailed comparisons for vertical tracks of meteorological fields and chemical
 23 species in the biomass burning (Fig. 9a) and dust events (Fig. 9b). For the biomass burning plume the 16th
 24 vertical profile on August 15th, 2016 near 20°S is shown while the 10th profile on August 17th, 2016 near
 25 25°N for the Saharan dust plume is shown. The comparison of the meteorological fields of temperature,
 26 virtual potential temperature, water vapor, relative humidity, wind speed and wind direction are shown in
 27 Fig. 8 and do not change between the different chemical options. The model forecasted temperature and
 28 virtual potential temperature almost overlap the ATom-1 measurements for both the August 15th and 17th
 29 vertical tracks. For water vapor and relative humidity, the variations of the vertical profiles are also
 30 reproduced by the model, except there are some smaller peaks in the observed profiles. There are still some
 31 differences between model and ATom-1 observations for wind speed and wind direction, which may be due
 32 to model vertical resolution. Overall, the model is able to capture the general vertical variations. For the
 33 chemical species (see Fig. 9), the modeled EC using GOCART scheme is almost identical to that by the
 34 RACM_GOCART scheme (the green line is overlapped by the blue line). Both EC concentration plots show
 35 a vertical variation of decreasing with altitude and the concentrations are overestimated above 2 km in
 36 biomass burning plume (see Fig. 9a) and above 4 km in dust storm (see Fig. 9b). The results using the
 37 RACM_SOA_VBS scheme shows much better performance to capture the vertical variations of EC. Other

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1 than a slight overestimation at 2-4 km biomass plume (see Fig. 9a first column), the EC vertical profile is
2 very consistent to that of the observation when using RACM_SOA_VBS scheme. In the biomass burning
3 event (see Fig. 9b first column), the modeled vertical profile with the RACM_SOA_VBS scheme captures
4 the general changes of the vertical variations much better than those of the GOCART and RACM_GOCART
5 schemes. As mentioned, previously, the assumption of no wet deposition for hydrophobic BC is the main
6 reason resulting in less EC at high altitude in the RACM_SOA_VBS scheme compared to the GOCART and
7 RACM_GOCART schemes. Due to less available observed data for sea salt, it is difficult to perform specific
8 comparisons, but both the observation and model show strong decreases with altitude. During the dust event
9 (see Fig. 9b third column), even though the modeled dust concentrations are lower at about 2-6 km than the
10 observed concentrations, they are close to the observation at the surface and upper levels. For the gas-phase
11 species, the model results are from GOCART_RACM (blue line) and RACM_SOA_VBS (red line) schemes.
12 The observed O₃ in the biomass burning event (see Fig. 9a fourth column) shows a peak at about 2 km height,
13 then it decreases with altitude, but increases again at about 5-9 km height. The model results from these two
14 schemes are quite consistent. They both indicate a slight enhancement at 1.5 km height, though it is not able
15 to capture the magnitude of the observed peak, which is underestimated by ~50%. For CO, the model can
16 reproduce the peak at about 2 km height very well, though it overestimates the mixing ratio by 25% below 1
17 km in the biomass burning event (see Fig. 9a 5th column). The detailed variations of the O₃ and CO vertical
18 profiles still show some slight differences between the model and observation, but the model generally
19 forecasts the vertical changes with altitude, and the CO using RACM_GOCART is slightly lower than that
20 of the RACM_SOA_VBS scheme above 5 km height.

21 4.2 Comparisons of aerosols and gas tracers between FIM-Chem and ATom-1 over the United States

22 Figure 10 shows the comparisons of EC and sulfate between ATOM-1 measurements and FIM-Chem model
23 with three different chemical schemes over the United States. Other than the underestimates of wet removal
24 for EC in GOCART and RACM_GOCART schemes that result in the overpredicted EC concentrations above
25 4 km height, the near surface (below 4 km) EC concentrations over southern California are also higher than
26 the observation. The overestimate over southern California is also shown in the RACM_SOA_VBS scheme.
27 Similarly, the predicted sulfate concentrations over southern California are much higher than the observation
28 too. Also, the surface sulfate concentrations throughout the U.S. are much higher than those of observations.

29 In the FIM-Chem model, the ~~anthropogenic~~ emissions are from the HTAP v2.1 inventory, which based on
30 the NEI2005 over United States. However, the BC emissions have declined by 50% in California from 1980
31 to 2008 following a parallel trend the reduction of fossil fuel BC emissions [Bahadur et al., 2011]. The older
32 emission inventory with relatively higher anthropogenic emissions of BC and SO₂ may possibly induce the
33 overestimates of near- surface BC and sulfate concentrations for the 2016 simulation in the model results
34 over southern California and other areas. To test this hypothesis we performed the same GOCART
35 retrospective experiment using the Community Emissions Data System (CEDs) anthropogenic emission
36 [Hoesly et al., 2018] instead of the HTAP v2.1 inventory. The CEDs anthropogenic emission is much

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1 stronger than HTAP over California for SO₂ (see Fig.11). Thus, a significant enhancement in sulfate
2 concentration near the surface of California is seen when using CEDS emissions, as shown in Figure 12. For
3 the sulfate concentrations at upper levels, the GOCART scheme (see Fig. 10b the second column) using the
4 background fields of H₂O₂, OH and NO₃ shows much better performance in capturing the relatively lower
5 sulfate at upper levels compared to the other two schemes.
6 Figure 13 shows the comparisons of OH and H₂O₂ in GOCART, RACM_GOCART and RACM_SOA_VBS
7 with ATom-1 observations. It can be seen that the prescribed OH is close to the ATom-1 observation, which
8 may be the major factor contributing to better sulfate agreement in GOCART. Considering the sulfur
9 chemical reaction mechanism and the aerosol scheme in RACM_SOA_VBS is completely different to that
10 in GOCART and RACM_GOCART, the comparison of oxidants may not be the only reason causing the
11 differences, which needs further analysis. For the gas species we compare CO, HCHO and O₃ (see Fig. 14)
12 using the RACM_GOCART and RACM_SOA_VBS schemes with the observation. Generally, the model
13 cases using either RACM_GOCART or RACM_SOA_VBS scheme show good performance in capturing
14 the CO and HCHO mixing ratios both at the surface and in the free troposphere. But they are both higher
15 than the observations near the surface over southern California, similar to EC and sulfate concentrations. This
16 may be also associated with the overestimation of anthropogenic emissions in the NEI-2005 over United
17 States for the year of 2016. Since CO and HCHO are precursors for O₃ production, the simulated O₃ also
18 shows slight enhancements compared to the observations that may be due to the higher CO and HCHO. Other
19 than that, the model is able to reproduce the O₃ profile over the US reasonably well, including the O₃
20 stratospheric intrusions at the upper levels. The simulated H₂O₂ in both RACM_GOCART or
21 RACM_SOA_VBS schemes show better agreement with the observations at the upper levels than the
22 prescribed H₂O₂ fields in GOCART (Fig. 13). While the much lower H₂O₂ near the surface in the
23 RACM_SOA_VBS may be associated with better O₃ performance near the surface (Fig. 13).
24 Figure 15 focuses on the 4th vertical profile over Kansas on August 23rd, 2016. The model results with
25 different chemical schemes are very consistent in simulating the meteorological fields. The modeled
26 temperature and virtual potential temperature show nearly exact agreement with the observations. But there
27 are still some shortcomings in forecast water vapor and relative humidity, especially above 6 km, where the
28 model results are overpredicted by nearly a factor of 2 and with less vertical variability. The vertical trend of
29 modeled wind speed and wind direction are close to the observed changes that increase with altitude. Similar
30 to Figure 9, the EC vertical profile using the RACM_SOA_VBS scheme, without the hydrophobic
31 assumption in wet removal, is similar to that of the observations while the other 2 schemes significantly
32 overpredict. Both the observations and models show decreasing vertical trend for sea salt and dust. The
33 GOCART scheme is able to reproduce the sulfate, except for the underestimate at 1.5-3 km. Otherwise, it
34 almost overlaps the observed profile at the upper levels. The O₃ vertical profile is reproduced by the model
35 using both RACM_GOCART and RACM_SOA_VBS schemes except a slight peak near 9 km where the
36 model is not able to capture the enhanced variability. The CO measurements have more fluctuations, but the
37 model roughly shows the major features of the vertical changes with altitude.

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1 5 Correlations between FIM-Chem model and ATom-1 observations

2 For the aerosol size range of the GOCART scheme, the PALMS dataset allows for model evaluation of the
3 default sea salt emission algorithms by summing those bins less than 3 μm in the model results. The
4 comparison between the GOCART forecasts and ATom-1 data for all sea salt observations below 6 km are
5 shown in Figure 16. Different colors show different flight dates from August 15th (blue dots), 17th (green
6 dots), 20th (orange), 22nd (red) and 23rd (purple). Generally, modeled sea salt appears too high, especially on
7 flights of August 15th (blue dots), 20th (orange dots) and 23rd (purple dots) above ~4km. Some high values
8 below ~4km are reproduced by the models on the flight of August 17th (green dots). Some of the disagreement
9 may be due to uncertainties in the size range of sea salt observations, particularly the upper cutoff of 3 μm
10 that is approximate (Murphy et al., 2019).

11 We also investigate the relationships of some key species for the biomass burning plumes observed on
12 8/15/17 and 8/17/17 between 22°S to 22°N below 6km (Fig. 17) for the RACM_SOA_VBS scheme. The
13 color bar indicates the latitude from south to north. Relative to CO, the model biomass burning emission
14 ratios are reasonable for EC with the modeled ratio (black color dots) somewhat lower than the observations
15 (color dots). We note that in Fig.6, O₃ in the biomass burning region for the RACM SOA VBS scheme is
16 underpredicted. To analyze this O₃ bias in more detail, scatter plots of modeled and observed NO_y versus CO
17 and O₃ versus NO_y between 22°S and 12° N below 6km altitude are shown in Fig.17b and Fig. 17d,
18 respectively. The observations in Fig. 17d show a much different, and better defined slope of O₃ versus NO_y
19 compared to the model using RACM SOA VBS scheme. NO_y, which is emitted entirely as NO_x in fresh
20 plumes, is much higher in the model, suppressing OH (e.g., Fig. 7), HO₂, and subsequent ozone formation.
21 The NO_y to CO ratios in Fig. 17b show evidence in the model of NO_y removal through HNO₃ scavenging,
22 but it's clear the NO_y (or NO_x) to CO emission ratio is too high in the fire emissions. The CO emissions
23 themselves appear too high (as also shown in Fig. 6). Other factors, such as VOC emission ratios or photolysis
24 effects from convective clouds may come into play, but these emission overestimates appear to put the
25 biomass burning region in a different photochemical regime than shown in the ATom-1 observations.

26 6 Conclusions

27 A two way fully inline coupled global weather -chemistry prediction model FIM-Chem has been developed
28 at NOAA Global Systems Laboratory (GSL) to forecast the chemical composition and quantify the impacts
29 on NWP. Three different gas/aerosol chemistry schemes - GOCART, RACM_GOCART and
30 RACM_SOA_VBS from WRF-Chem have been implemented into FIM-Chem with some modifications as
31 different options of chemical schemes. The major conclusions are summarized as follows:
32 First, the RACM GOCART mechanism with explicitly simulated H₂O₂, OH and NO₃ is compared to the
33 base GOCART mechanism having a simple parameterization of sulfur/sulfate chemistry using prescribed
34 background fields of OH, H₂O₂ and NO₃. The explicit treatment results in about 10 $\mu\text{g}/\text{m}^3$ reductions of
35 sulfate and 15 $\mu\text{g}/\text{m}^3$ of PM_{2.5} over the eastern US, as well as more than 20 μg reductions of PM_{2.5} over eastern

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1 China. Meanwhile, the simulated instantaneous H₂O₂ is lower by 20% over eastern US and 40-60% over
 2 India and eastern Asia, while the OH is 80% lower over eastern China in the RACM_GOCART scheme.
 3 In this study, the evaluation and analysis of model performance are focused on the fire events over the Eastern
 4 Atlantic from south to north on August 15th and 17th 2016, and the flight over the United States from
 5 Minnesota to southern California using the NASA ATom-1 observations.
 6 For the evaluation over Atlantic, the GOCART and RACM_GOCART results are very consistent in
 7 forecasting sulfate, sea salt and EC due to the same aerosol mechanism. For the fire events sampled near the
 8 equatorial Atlantic (e.g. Fig. 6), the GOCART and RACM_GOCART schemes show good performance in
 9 reproducing the profiles of EC, and CO is captured reasonably well with the RACM_GOCART and
 10 RACM_SOA_VBS schemes. Generally, EC is simulated well by GOCART and RACM_GOCART
 11 mechanisms up to 4 km but above this the mechanisms are biased high, while EC in the
 12 RACM_SOA_VBS scheme shows much better performance than that of the GOCART and
 13 RACM_GOCART schemes at the upper levels. This is because it's assumed there is no wet deposition for
 14 hydrophobic BC in the GOCART and RACM_GOCART schemes, which results into an underestimate of
 15 EC wet removal and overestimate of EC concentrations at higher levels. The CO mixing ratio above ~2 km
 16 is underestimated over the tropics and overestimated at altitudes below ~2km, which may be related to lower
 17 simulated fire injection heights in the model. Otherwise, the general CO profiles are well reproduced. Both
 18 RACM_GOCART and RACM_SOA_VBS schemes are able to consistently reproduce O₃ mixing ratios,
 19 including the stratospheric intrusion above ~9 km at 40°S. There is some slight underestimation of O₃ near
 20 the tropics, which might be associated with the underprediction of CO outside the biomass burning signature
 21 region. We also evaluated other gas-phase species: HCHO, OH and H₂O₂, which are important precursors to
 22 many other chemical species within the RACM_GOCART and RACM_SOA_VBS schemes (see Fig. 7).
 23 Generally, the pattern of the modeled HCHO, OH and H₂O₂ mixing ratio are almost the same as that of the
 24 ATom-1 observations except for some underestimates above 9 km for HCHO and OH at some latitudes, and
 25 some overestimates of H₂O₂ above 6 km in the southern hemisphere.
 26 For the evaluation from Minnesota to southern California, all of the chemical schemes are able to reproduce
 27 the general vertical gradients seen in the observations. The RACM_SOA_VBS scheme is able to reproduce
 28 the vertical profile of EC much better than that of the GOCART and RACM_GOCART schemes, which
 29 overestimate the EC concentrations above 2-4 km due to the assumption of no wet deposition for hydrophobic
 30 BC. This comparison highlights the value of the ATom-1 data in examining basic assumptions within the
 31 wet removal parametrization of carbonaceous aerosol in the GOCART mechanism. The high SO₂ emissions
 32 from either anthropogenic or fire sources play important role in enhancing the sulfate production. There are
 33 high biases above ~3km for sulfate in the RACM_GOCART and RACM_SOA_VBS schemes. Results from
 34 the RACM_GOCART and RACM_SOA_VBS schemes show consistency with observed O₃ and CO vertical
 35 profiles during the fire events. Both schemes show a slight enhancement of O₃ at 1.5 km even though it
 36 underestimates the magnitude of the observed peak. For CO, the model results capture the peak at about 2
 37 km very well but overestimates the mixing ratio by about 30% near the surface. For the gas-phase species,

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1 the model either using the RACM_GOCART or RACM_SOA_VBS scheme shows very good ability in
2 forecasting the CO, O₃ and HCHO mixing ratio both at the surface and free troposphere, including the O₃
3 stratospheric intrusions at the upper levels (Fig. 14). For CO, a precursor for O₃ production, there appears to
4 be overestimated emissions over California causing much higher surface mixing ratios in the forecasts than
5 observed. For the comparisons of vertical profiles over California on August 23rd 2016, the modeled
6 meteorological fields of temperature and potential temperature show agreement with the observations. The
7 modeled water vapor and relative humidity are consistent with observations below 6 km though they are
8 overestimated above 6km. The RACM_SOA_VBS scheme shows the best agreement with EC. For sulfate,
9 the GOCART scheme is almost the same as the observation above 3km while it overestimates near the surface
10 due to the high anthropogenic emissions used within the inventory. The simulated O₃ and CO vertical profiles
11 almost overlap the ATom-1 measurements but with less vertical variability. Though data is somewhat sparse
12 in our analysis, the sea salt emission algorithm appears to be a model component that could be improved due
13 to apparent consistent overestimation.

14 The scatter plots of sea salt and gas tracers from biomass burning plumes shows that modeled sea salt appears
15 too high and some of the disagreement may be due to uncertainties in the size range of sea salt observations
16 (Fig. 16), and the NO_x (or NO₂) to CO emission ratio is too high in the fire emissions (Fig. 17). These emission
17 overestimates may put the biomass burning region in a different photochemical regime than shown in the
18 ATom-1 observations.

19 The comparison in this study successfully demonstrates that the FIM-Chem model with three difference
20 chemical schemes show good performance in forecasting the chemical composition for both aerosol and gas-
21 phase tracers when compared with the high temporal resolution (1-second) observations of ATom-1. The wet
22 removal assumption for hydrophobic BC is not reasonable, which needs to be improved in the GOCART and
23 RACM_GOCART schemes. It is not necessary to use the complexity of a gas-phase scheme if the focus is
24 only on aerosol forecasts, in order to save time and computer resources. Using anthropogenic emissions for
25 the specific year of the simulation may help to improve the forecasts. Also, a new dynamic core of Finite-
26 volume cubed-sphere dynamical core (FV3) developed by GFDL will be used to replace of FIM and coupled
27 with the chemical schemes in the next generation global prediction system (NGGPS), as FV3GFS-Chem, by
28 using that to demonstrates the chemical impacts on NWP.

30 Code and data availability

31 Basically, the chemical modules of GOCART, RACM_GOCART and RACM_SOA_VBS are based on the
32 WRF-Chem 3.7, which can be obtained from
33 http://www2.mmm.ucar.edu/wrf/users/download/get_source.html. The FIM-Chem v1 code and model
34 configuration for chemical composition forecast here are available at <http://doi.org/10.5281/zenodo.5044392>.
35 ATom-1 data is publicly available at the Oak Ridge National Laboratory Distributed Active Archive Center:
36 https://daac.ornl.gov/ATOM/guides/ATOM_merge.html (Wofsy et al., 2018).

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1 **Author contribution**

2 Li Zhang and Georg A. Grell developed the model coupling code and implemented the chemical modules
3 from WRF-Chem into FIM model. Li Zhang designed the experiments and performed the simulations. Stuart
4 A. McKeen evaluated the model performance and provided the suggestions to improve model performance.
5 Ravan Ahmadov developed the RACM-SOA-VBS scheme in WRF-Chem. Karl D. Froyd and Daniel
6 Murphy performed the measurements and provided the measured data of ATom-1 experiments. Li Zhang
7 prepared the manuscript with contributions from all co-authors.

8 **Competing interests**

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

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1 Table 1. Chemical Scheme comparison.

	<u>GOCART</u>	<u>RACM GOCART</u>	<u>RACM SOA VBS</u>
<u>Number of transport Tracers</u>	19	68	103
<u>Number of Chemical Reactions</u>	4	214	233
<u>Aerosol scheme</u>	<u>GOCART</u>	<u>GOCART</u>	<u>SOA VBS</u>
<u>GAS-phase chemistry scheme</u>	/	<u>RACM</u>	<u>RACM</u>
<u>Computational expense of 24 hours forecast</u>	<u>~4 minuets</u>	<u>~19 minuets</u>	<u>~22 minuets</u>

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1 **Figure captions.**

2 **Figure 1.** Vertical profiles and transect time series of the ATom-1 flight tracks on August 15th and 17th, 2016
3 over Atlantic Ocean and August 23rd 2006 over US.

4 **Figure 2.** ~~5-days forecast started from 00:00 UTC July 29th 2016 of surface PM_{2.5} and sulfate using (a)~~
5 GOCART and (b) RACM_GOCART schemes, and their (c) differences (RACM_GOCART minus
6 GOCART) at ~~00:00 UTC August 3rd 2016~~. Unit: $\mu\text{g}/\text{m}^3$.

7 **Figure 3.** Comparisons of ~~5-days forecast started from 00:00 UTC July 29th 2016 of surface H₂O₂, OH, and~~
8 NO₃ between (a) GOCART and (b) RACM_GOCART schemes, and their differences (RACM_GOCART
9 minus GOCART) at ~~(c) 00:00 UTC and (d) August 3rd 2016~~. Unit: ppb.

10 **Figure 4.** ~~5-days forecast started from 00:00 UTC July 29th 2016 of surface O₃ using RACM_GOCART~~
11 scheme at ~~12:00 UTC August 2nd and 00:00 UTC August 3rd 2016~~. Unit: ppb.

12 **Figure 5.** ~~5-days forecast started from 00:00 UTC July 29th 2016 of surface SOA using RACM_SOA_VBS~~
13 scheme at ~~12:00 UTC August 2nd and 00:00 UTC August 3rd 2016~~. Unit: $\mu\text{g}/\text{m}^3$.

14 **Figure 6.** Height-latitude profiles of EC, CO and O₃ over Atlantic on August 15th and August 17th, 2016 for
15 (a) ATom-1; (b) RACM_GOCART; and (c) RACM_SOA_VBS.

16 **Figure 7.** Height-latitude profiles of HCHO, OH and H₂O₂ over Atlantic on August 15th and August 17th,
17 2016 for (a) ATom-1 observations; (b) RACM_GOCART; and (c) RACM_SOA_VBS.

18 **Figure 8.** ATom-1 observations and model results for temperature, virtual potential temperature, water vapor,
19 relative humidity, wind speed and wind direction in the (a) biomass burning and (b) dust events. The biomass
20 burning plume is from August 15, 2016, profile #16 near 20°S while the Saharan dust plume is from August
21 17, 2016, profile #10 near 25°N.

22 **Figure 9.** Comparisons between ATom-1 observations and model vertical profiles of EC, sea salt, dust, O₃
23 and CO in ~~(a) biomass burning event and (b) dust storm event~~. The biomass burning plume is from August
24 15, 2016, profile #16 near 20°S while the Saharan dust plume is from August 17, 2016, profile #10 near 25°N.
25 Green and blue lines are nearly identical for aerosol.

26 **Figure 10.** Height-latitude profiles of EC and sulfate over United States on August 23rd, 2016 for (a) ATom-
27 1; (b) GOCART; (c) RACM_GOCART; and (d) RACM_SOA_VBS.

28 **Figure 11.** ~~Anthropogenic emissions of SO₂ of (a)HTAP and (b) CEDS inventories on August~~. Unit:
29 $\text{mol}/\text{km}^2/\text{hour}$.

30 **Figure 12.** ~~Height-latitude profiles of sulfate over United States on August 23rd, 2016 for (a) ATom-1, (b)~~
31 ~~GOCART with HTAP, (c) GOCART with CEDS anthropogenic emission.~~

32 **Figure 13.** ~~Height-latitude profiles of OH and H₂O₂ over United States on August 23rd, 2016 for (a) ATom-~~
33 ~~1; (b) GOCART; (c) RACM_GOCART; and (d) RACM_SOA_VBS.~~

34 **Figure 14.** ~~Height-latitude profiles of CO, O₃ and HCHO over United States on August 23rd, 2016 for (a)~~
35 ~~ATom-1; (b) RACM_GOCART; and (c) RACM_SOA_VBS.~~

36 **Figure 15.** Observations and model results for profile #4, 8/23/16 over southeastern Kansas.

37 **Figure 16.** GOCART model forecast versus ATom-1 observed sea salt below 6 km.

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1 **Figure 17.** Model (black color dot) and observation (color dot) ratios of (a) EC relative to CO; (b) NO_y
2 relative to CO; (c) O₃ relative to CO and (d) O₃ relative to NO_y. Color scale is degree latitude.
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