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Scoping Study for Development/Application of CAMx for the Northern Hemisphere

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EXECUTIVE SUMMARY

The TCEQ uses the Comprehensive Air quality Model with Extensions (CAMx) for State Implementation Planning (SIP) purposes. CAMx uses boundary conditions to specify air quality at the perimeter of the CAMx model domain and the TCEQ uses a global chemical transport model (GEOS-Chem) to develop CAMx boundary conditions for a domain that spans much of North America. However, GEOS-Chem is primarily intended for studies of the global atmosphere, and so uses different atmospheric chemical mechanisms than CAMx and lacks a source apportionment capability for tracking ozone and particulate matter (PM) contributions back to the source categories and regions where precursors were emitted. These issues could be eliminated by developing a capability to run CAMx on a domain covering the entire Northern Hemisphere.

This report presents a scoping study to develop the foundation for a CAMx hemispheric modeling platform. Based on a literature review of current hemispheric and global modeling systems, we investigate approaches and identify technical hurdles related to configuring and running CAMx for the entire northern hemisphere. From these reviews, we develop recommendations for the CAMx hemispheric modeling platform that consider data needs and data sources, horizontal and vertical grid structures, how to interface the hemispheric grid with TCEQ's regional CAMx grid, and needed model updates. Developing a CAMx hemispheric modeling platform would provide a consistent, stream-lined global-to-regional modeling system that would simplify the TCEQ's modeling procedures and add important Probing Tools such as Source Apportionment across the northern hemisphere.

Guided by information and recommendations from this scoping study, we have developed a recommended approach, estimated cost and schedule, for a "proof-of-concept" implementation of CAMx at the hemispheric scale, including necessary software development and testing. The approach includes a list of model and pre-processing updates and improved data sources needed to better address chemical and physical processes at the global scale.

1.0 INTRODUCTION

1.1 Background

The TCEQ uses the Comprehensive Air quality Model with Extensions (CAMx; Ramboll, 2018) for State Implementation Planning (SIP) purposes. CAMx requires input datasets that specify the temporal and spatial distributions of a multitude of chemical concentrations for initial and boundary conditions. TCEQ uses a global chemical transport model called the Goddard Earth Observing System model with Chemistry (GEOS-Chem; Bey et al., 2001; Harvard, 2018) to develop CAMx initial/boundary conditions on a domain spanning much of North America. However, GEOS-Chem is primarily intended for studies of the global atmosphere and so uses different atmospheric chemical mechanisms than CAMx to simulate ozone and particulate matter (PM). Additionally, GEOS-Chem lacks a source apportionment capability for tracking ozone and particulate matter (PM) contributions back to the source categories and regions where precursors were emitted. Finally, due to different priorities, GEOS-Chem's development and science updates are frequently out-of-step with the latest enhancements to CAMx.

These issues could be eliminated by developing a capability to run CAMx on a domain covering the entire Northern Hemisphere. The advantages and tangible benefits of hemispheric CAMx applications include: (1) a source of chemically consistent CAMx boundary conditions for TCEQ's regional modeling applications; (2) a seamless integration of the CAMx Source Apportionment tool to track contributions from foreign sources to ozone and PM in Texas and other states; (3) chemical and physical consistency between global and regional scales in a single model framework; and (4) immediate availability of future CAMx updates and enhancements to global applications without the need to implement them in third-party global models. Since TCEQ is familiar with CAMx and its Source Apportionment features, a hemispheric configuration of CAMx could be readily subsumed into TCEQ's set of modeling tools.

1.2 Purpose and Objectives

This Work Order comprises a scoping study to develop the foundation for a CAMx hemispheric modeling platform. Based on a literature review of current hemispheric and global modeling systems, we investigate approaches and identify technical hurdles related to configuring and running CAMx for the entire northern hemisphere. From these reviews, we have gained insights into data needs and data sources, horizontal and vertical grid structures, needed technical updates, and how to interface the hemispheric grid with TCEQ's regional grid. This report concludes with a recommended approach for an initial proof-of-concept hemispheric CAMx application as a future project, as well as a prioritized list of model and pre-processing updates and improved data sources needed to better address chemical and physical processes at the global scale.

2.0 MODELING CONSIDERATIONS

The US Environmental Protection Agency (EPA) has also recognized the value of extending regional photochemical grid models to hemispheric scales. A recent paper by Mathur et al. (2017; hereafter M17) documents their work with the Community Multiscale Air Quality (CMAQ) model (Appel et al, 2017). It lays the groundwork for the same data requirements and technical issues that we face with CAMx, and the authors have expended considerable effort with respect to gathering emissions data, meteorological modeling, and treating stratospheric ozone. Therefore, our review relies heavily on the comprehensive developments reported by M17.

The sub-subsections below discuss and consider issues related to model configuration, available supporting datasets, and necessary CAMx modifications that would be needed for hemispheric applications. This section concludes with a summary of how regional model initial and boundary conditions would be derived from hemispheric output fields, and how Source Apportionment would be coupled between global and regional applications.

2.1 Model Configuration

2.1.1 Grid Projection and Resolution

Full global chemical transport models run on a variety of specialized grid structures that alleviate the spatial singularities that occur at the poles. Often these grids are matched to the driving general circulation models to which they are coupled (e.g. AM3; Donner et al. 2011), or to the structures of historical global meteorological analyses (e.g., GEOS-Chem; Harvard, 2018). Global horizontal resolutions can range from ~0.5 degrees (~50 km) to 2 or 3 degrees (200-300 km). Vertical grids can approach 100 layers, but because these models extend to enormous altitudes, their vertical resolution within the troposphere is typically similar to, or somewhat coarser than, common regional photochemical model applications. Because of their specialized, global-focused configuration, the horizontal grid and vertical layer structures employed in full global models are either incompatible or inappropriate choices for a hemispheric-extended photochemical model.

M17 utilized CMAQ's polar stereographic (PS) projection option for their hemispheric configuration, which is consistent with that used in the Weather Research and Forecasting (WRF; Skamarock et al., 2008) meteorological model with which CMAQ is intimately coupled. The PS projection is the only option available in WRF and CMAQ that can include the North Pole without a mathematical singularity. CAMx also includes the capability to match several WRF projections, including PS. As a limited-area model, all WRF domains are defined as a rectangular set of grid points on the projection plane. This means that hemispheric PS WRF domains are not bounded by a circle defined at a particular latitude (such as a radar scope, the most common way PS maps are presented). In a PS projection, the "north-south" direction of the rectangular grid (i.e., the definition of orthogonal *u* and *v* wind components) is defined to align along a particular meridian of the user's choice, and the domain corners extend farther south than the domain sides. Additionally, this means that a North-South *v*-component wind at the anchor meridian will rotate to an East-West wind component 90 degrees of longitude from that point (and vice-versa for the *u*-component). This makes mapping grid cells and determining their compass orientation a tricky exercise.

Like any photochemical model application, domain coverage and horizontal/vertical resolution must be balanced against runtimes and computer costs. We anticipate that a typical hemispheric model application would simulate a year or more, depending on several factors including the necessary “spin-up” time for the model to reach chemical equilibrium from initial conditions. For practical purposes, an annual hemispheric application should be able to complete in a few days to a week of “wall-clock” time using optimal parallelization.

Horizontally, the domain should extend to equatorial latitudes so that large-scale circulation features are entirely captured. A PS projection tangent at the North Pole results in increasing spatial distortion away from the pole with excessive distortion at the Equator. However, the WRF PS projection employs a secant latitude; i.e., a constant latitude ring where the PS projection intersects the sphere and represents “true” distances, much like the “true latitudes” of a Lambert Conformal projection. This reduces spatial distortions for domains that extend to the equator.

Horizontal resolution should be equivalent or better than the global models currently employed to provide regional boundary conditions (e.g. 200-300 km), and should result in a total number of grid cells that are consistent with regional applications to maintain manageable memory requirements and runtime efficiency (e.g., 200x200). Horizontal resolution is a key factor in properly simulating intercontinental transport (and thus foreign source apportionment), as elevated plumes transported in the free troposphere are overly diffused in coarse grids. Eastham and Jacob (2017) investigated this effect using the GEOS-Chem global chemistry model by varying horizontal resolution from 0.25°×0.3125° (~25-30 km) to 4°×5° (~400-500 km) and holding the vertical layer structure fixed (72 standard GEOS-5 meteorological layers from the surface to ~80 km). They found that intercontinental plume coherence was better preserved by increasing horizontal resolution to about 1°×1° (~100 km) but did not see improvements at higher resolutions. Eastham and Jacob (2017) hypothesized that the native vertical resolution in GEOS-5 meteorology was limiting further progress in preserving the structure of intercontinental plumes by more than a few days.

Adequate vertical resolution is important to resolve boundary layers (particularly extensive shallow marine layers), free tropospheric transport, and stratosphere-troposphere exchange (STE) of ozone. Zhuang et al. (2017) recently conducted a follow-on study to the Eastham and Jacob (2017) paper and confirmed that both horizontal and vertical resolution impact three-dimensional numerical diffusion of intercontinental transport plumes in the free troposphere. They have recommended an optimal $\Delta x/\Delta z$ ratio ranging from 700 to 1500, which in their tests preserved plume coherence up to a week or more. The 72-layer structure used in the work of Eastham and Jacob (2017) resolved the mid-troposphere at ~500 m, resulting in $\Delta x/\Delta z \approx 200$ at 1° horizontal resolution. As point out by Zhuang et al. (2017), this was the major limitation to further reductions in plume diffusion below 1° noted by Eastham and Jacob (2017); at such horizontal resolution, vertical layers should be at most ~150 m in the mid-troposphere.

M17 employed a grid structure that appears to satisfy most of the requirements and issues discussed above. Identical projections and grids were employed for both WRF and CMAQ to maximize consistency and limit error related to interpolation and averaging to different structures. Their PS domain comprises a 187x187 grid with 108 km resolution (~1°). They ran with both 35 and 44 layers up to a model top at 50 mb (18-20 km). Their tests revealed that the 35 layer structure was overly diffusive, particularly around the tropopause, leading to poor agreement with upper tropospheric ozone profiles. M17 found much improved performance using 44 layers, and like Zhuang et al. (2017), recommend the use of as many layers as practical to resolve the free troposphere and lower stratosphere. Like GEOS-Chem, M17's 44 layers resolve the mid-troposphere with layers depths typically ~500 m ($\Delta x/\Delta z \approx 200$). Tests with additional layers would be necessary to confirm any improvement to the coherence of intercontinental transport plumes on the M17 grid.

2.1.2 Tropospheric Chemistry

Global-scale photochemical models must properly characterize tropospheric chemistry over a large range of spatial and temporal scales, ranging from faster reactions among hundreds of precursor species within urban/industrial source areas to slower reactions involving long-lived reservoir species during long-range transport. At the same time, chemical mechanisms must be efficient for the global models to be useful and practical. M17 tested two chemical mechanisms: CB05TU was used for most hemispheric applications, and RACM2 (Sarwar et al., 2013) was tested and compared to CB05TU for a summer period in 2006. CB05TU includes EPA updates in the treatment of organic nitrates (important as reservoir species), a new large halogen chemistry mechanism (important oceanic source of radicals for photochemistry and ozone depletion), and updated ozone deposition rates to ocean surfaces. Both CB05TU and RACM2 are tied into CMAQ's AERO6 PM chemistry, which includes RADMv2 aqueous chemistry, ISORROPIAv2 inorganic partitioning, and a comprehensive organic aerosol scheme.

The most recent Carbon Bond mechanism available CAMx (CB6r4) includes improvements to organic nitrate chemistry and includes halogen reactions for oceanic emissions and subsequent products. CB6r4 includes a reduced halogen mechanism that considers only inorganic iodine to improve efficiency while maintaining the most important halogen influences on ozone chemistry in maritime environments. CB6r4 is tied into the "CF" aerosol chemistry, which uses RADMv1.5 for aqueous chemistry, SOAP for organic chemistry and partitioning, and ISORROPIA or EQSAM for inorganic partitioning. Our tests with the new EQSAM approach show that it is slightly faster than ISORROPIA (6-8%), results in very similar PM production and spatial/temporal patterns, yet removes numerical artifacts that can be generated by ISORROPIA. Overall, CB6r4/CF is appropriate to simulate chemistry at the wide range of spatial and temporal scales needed for hemispheric photochemical applications. The use of EQSAM may be preferable over ISORRPIA to improve model speed.

2.2 Available Datasets

2.2.1 Meteorological Inputs

Generally, global models employ routine, publicly-available global meteorological analyses (for historical simulations), or global climate model (GCM) output (for future simulations). For example, the GEOS-Chem global chemistry model uses historical Goddard Earth Observing System (GEOS) datasets, while the Model for Ozone and Related Chemical Tracers (MOZART) uses historical NCEP/NCAR-reanalysis meteorological fields, and its replacement the Whole Atmosphere Community Climate Model (WACCM) runs 10-day forecasts driven by NASA/GMAO GEOS-5 meteorology. These global analysis datasets likely provide sufficient variable fields to be a viable source of meteorological data for hemispheric CAMx applications. This would add flexibility in using off-the-shelf analyses for any historical year (e.g., GEOS or NCEP/NCAR), short-term forecast cycle (e.g., WACCM/GEOS-5), or distant future conditions (e.g., GCM).

However, a new meteorological interface program would need to be developed to translate global analysis fields to the hemispheric polar CAMx grid and to the specific input variables required by CAMx. Also, given some mass consistency issues inherent in the global analyses, most global models must employ various internal adjustments as they run, such as vertical "pressure fixers", to ensure that their three-dimensional transport solutions are mass conservative and consistent. While this may not be an issue for CAMx, given its internal approach to calculate vertical fluxes in a mass-consistent manner, it would need to be carefully investigated, and if needed, a solution would need to be implemented in the new global analysis interface program. Additionally, momentum fields in global analyses do not necessarily match higher-resolution WRF-generated meteorological fields along the boundaries of the regional models. This is generally related to the coarse spatial and time resolution of the global analyses. This can lead to inconsistencies in source apportionment linkages between hemispheric and regional CAMx applications.

Therefore, to achieve maximum consistency, WRF should be run in a consistent manner for both global and regional applications. The meteorological coupling through WRF would limit errors and inconsistencies resulting from different projections, horizontal resolutions, and layer structures. Additionally, WRF can be configured to support model-specific processes within CAMx, such as specific fields to support sub-grid convection in the CAMx cloud-in-grid (CiG) sub-model (as described below).

M17 applied WRF on the hemispheric CMAQ grid system for the period of 1990-2010 (Xing et al., 2015). WRF was guided by 6-hour NCEP/NCAR Reanalysis fields at 2.5-degree horizontal resolution. The nudging fields were augmented with point measurements from the Automated Data Processing (ADP) global observation network. M17 configured WRF to use the Pleim-Xiu land surface model with indirect temperature and moisture nudging and global MODIS 20-category landcover data. They used the ACM2 boundary layer scheme and RRTMG to simulate short and longwave radiation fluxes. Xing et al. (2015) do not state which cloud microphysics or sub-grid convection schemes were employed.

Developmental/demonstration hemispheric CAMx applications could use M17's 1990-2010 WRF datasets directly, although we recognize that this period does not align with TCEQ's current modeling programs. Alternatively, WRF could be run and evaluated for a more pertinent period using a similar configuration to M17/Xing et al. (2015). Note that an appropriate CAMx spinup ranging from months to a full year may be necessary, so WRF would need to be run for up to 2 full years to support a hemispheric CAMx application.

Deep convection is an important process at global scales as it provides an efficient mechanism to loft emissions and secondary pollutants for long-range transport in the upper troposphere. It is also an important process for returning globally-transported pollutant back to surface, and for chemistry and wet removal. At the spatial resolutions envisioned for hemispheric CAMx applications (~100 km), sub-grid convection likely comprises a large fraction of total convective transport. Therefore, WRF should be configured with a specific sub-grid cloud scheme that supports the CAMx CiG option, which at this point is EPA's "scale-aware" version of the Kain-Fritsch scheme. The WRFCAMx interface programs could also easily be modified to use output from the scale-aware Grell convection scheme to increase CAMx flexibility.

2.2.2 Emission Inputs

2.2.2.1 Anthropogenic global emissions

The hemispheric CMAQ applications described by M17 have considered two primary sources of global anthropogenic emissions. Initial applications used the Argonne National Laboratory (ANL) global emissions inventory compiled for NASA's Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission. Subsequent applications have used year-specific estimates from the Emission Database for Global Atmospheric Research (EDGAR) version 4.2, which reports emissions for 17 anthropogenic sectors and large-scale biomass burning on a 0.1° x 0.1° resolution grid. The SMOKE model was updated to support hemispheric CMAQ applications using EDGAR.

In addition to these inventories, there are several other global anthropogenic emission inventories that could be considered for CAMx hemispheric applications. All are public databases and many of them are available from the Emissions of atmospheric Compounds and Compilation of Ancillary Data (ECCAD) database (http://eccad.aeris-data.fr), which is part of the Global Emissions InitiAtive (GEIA; previously known as Global Emissions Inventory Activity). Table 1 provides a list of current global inventories; this table was adapted from recent presentations by Clare Granier, the GEIA database manager, and Greg Frost, the GEIA co-chair.

The last entry in Table 1 is the Community Emissions Data System (CEDS). This global anthropogenic emissions inventory has been developed by the Joint Global Change Research Institute (JGCRI), a joint venture of Pacific Northwest National Laboratory (PNNL) and the University of Maryland for use in the Coupled Model Inter-comparison Project Phase 6 (CMIP6). The inventory is released through the Earth System Grid Federation (ESGF). The data system produces emission estimates by country, sector, and fuel with the following characteristics:

Table 1. List of candidate global anthropogenic emission inventories for hemispheric CAMx applications.

Inventory	Period	Notes
ACCMIP	1980-2000	Lamarque et al. (2010); http://eccad.aeris-data.fr
MACCity	1980-2010	Extension of the ACCMIP dataset; Granier et al. (2011); http://eccad.aeris-data.fr
EDGAR 4.3.1	1970-2010	Crippa et al. (2016); http://edgar.jrc.ec.europa.eu/overview.php?v=pegasos
HTAP_v2.2	2008 and 2010	Janssens-Maenhout et al. (2015); http://edgar.jrc.ec.europa.eu/htap_v2/
ECLIPSE V5	1990-2030	Stohl et al. (2015) and Klimont et al. (2017); http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5.html
CEDS	1750-2014	Hoesly et al. (2018); http://www.globalchange.umd.edu/ceds/

1. Annual estimates of anthropogenic emissions (not including open burning) to latest full calendar year over the entire industrial era; updated every year.
2. Emissions of aerosols (BC, OC), aerosol precursor and reactive compounds (SO_2, NOx, NH_3, CH_4, CO, NMVOC), and CO_2 (as reference).
3. State/province spatial detail for large countries – *in progress.*
4. Seasonal cycle (monthly) and aggregate NMVOCs by sector/sub-sector.
5. Gridded emissions (up to 0.1° resolution) w/ sub-national resolution for large countries.
6. Uncertainty estimated at the same level (country, fuel, sector) – *in progress.*

CEDS has replaced EDGAR 4.3.1 as the default inventory in GEOS-Chem v11-02 and later, although EDGAR emissions will be available as an option (http://wiki.seas.harvard.edu/geos-chem/index.php/CEDS_anthropogenic_emissions). For hemispheric CAMx applications, we recommend CEDS or EDGAR as sources of anthropogenic inventories. An approach similar to hemispheric CMAQ (using EDGAR) can be used to develop model-ready emissions for CAMx.

2.2.2.2 Natural global emissions

Important natural emission categories for hemispheric applications include biogenic emissions, lightning NOx, methane emissions, windblown dust, sea salt, and halogenated very short-lived substances (VSLSs) from the oceans (marine emissions), such as halomethanes and inorganic halogens. These halogenated species break down quickly and are the major contributors of bromine and iodine to the atmosphere. As discussed in Section 2.1.2, these halogens are an important source of radicals for photochemistry in the marine atmosphere.

The hemispheric CMAQ applications described by M17 included biogenic VOC emissions and lightning NOx emissions obtained from the GEIA database. Monthly biogenic emissions from GEIA were resolved to hourly values for each day, while monthly lightning NOx emissions were uniformly distributed to hourly values. GEIA data may be adequate for initial “proof-of-concept” CAMx applications at hemispheric scales, but ultimately we recommend using MEGAN since the GEIA biogenic emissions are considered to be obsolete (e.g., http://wiki.seas.harvard.edu/geos-chem/index.php/Biogenic_emissions). For lightning NOx emissions, we would ultimately recommend using a new preprocessor that Ramboll will develop in 2018 that can utilize information from WRF on convective cloud/rainfall activity.

M17 describe updates to the CMAQ in-line sea-salt module that include a parameterization to estimate marine emissions of bromine and iodine-containing compounds for three categories (halocarbons, inorganic bromine, and inorganic iodine). The halocarbons include five bromocarbons ($CHBr_3$, CH_2Br_2,CH2BrCl, CHBrCl2, $CHBr_2Cl$) and four iodocarbons (CH_3I, CH_2ICl, CH_2IBr, CH_2I2). The halocarbon emissions are estimated using monthly average climatological chlorophyll-a (Chl-a) concentrations derived from the Moderate Resolution Imaging Spectroradiometer (MODIS). A similar approach is available in the CAMx SEASALT preprocessor. SEASALT can support hemispheric applications by supplying halomethane and inorganic halogen emissions (iodides, bromides, chlorides) in the case that the full CB6r2h halogen mechanism is used, or just inorganic iodine emissions in the case that CB6r4 is used.

M17 describe updates to the CMAQ windblown dust emission parameterization for global applications, including an improved map of erodible areas and reduced threshold friction velocity to lift dust from the surface. Future versions of hemispheric CMAQ will include a new physics-based windblown dust emission parameterization (Foroutan et al., 2017). Hemispheric CAMx applications should use Ramboll's new windblown dust pre-processor, which has recently been distributed to TCEQ for testing. Ramboll's approach is based on the global EMAC (ECHAM/MESSy) chemistry-climate model (Klingmueller et al., 2017; Astitha et al., 2012).

Emissions of NO from soil or SO_2 from volcanos were not considered in the CMAQ hemispheric applications. Soil NO could be addressed in hemispheric CAMx applications if biogenic emissions are derived using MEGAN. Volcanic SO_2 emissions could be included based on satellite data that quantify annual average SO_2 emissions from major volcanoes in recent years.

2.2.3 Initial/Boundary Conditions

Hemispheric models require boundary conditions along their equatorial boundaries. M17 applied "clean" lateral boundary profiles for their hemispheric CMAQ applications. Based on the argument of a "1-year exchange time" between the southern and northern hemispheres, M17 states that impacts from lateral boundary conditions are generally confined to lower latitudes with little propagation into domain. It is well documented that the intertropical convergence zone (ITCZ) effectively restricts air exchange between the hemispheres. Note that CMAQ does not define top boundary conditions, so pollutant distributions near the top of the model may be dependent somewhat on lateral boundary conditions, but mostly on how the simulation evolves via three-dimensional transport throughout the hemisphere.

Both hemispheric and global models require initial conditions, which usually define a relatively simple/clean state. The models rely on extensive "spin-up" periods of a year or more to reach a chemically mature characterization of the global atmosphere. Indeed, M17 applied the same clean profiles for both lateral boundary conditions and initial conditions, and ran a 12-month spin-up period prior to the actual simulation year of interest. The standard approach to run the GEOS-Chem global model also involves running a spin-up period of at least 6 months to a year.

Hemispheric CAMx applications could use similar simple/clean lateral boundary and initial conditions, with the requisite spin-up period. However, existing global model output (e.g., the on-line MOZART library or GEOS-Chem runs developed for TCEQ) could be utilized to specify initial and boundary (lateral and top) conditions just as for regional modeling. This would remove the arbitrary characterization of near-equatorial profiles, provide valuable information at the top of the model, and dramatically shorten spin-up times from initial conditions to perhaps one month to reach a hemispheric "equilibrium". Special considerations for top boundary conditions are needed to address stratospheric ozone. These are discussed in the next section.

2.3 CAMx Model Updates

2.3.1 Stratospheric Ozone

Stratospheric ozone contributes 20-50% of the tropospheric ozone column (Lelieveld and Dentener, 2000) with large temporal and spatial variability. This fraction is increasing as a result of precursor emission reductions and climate change (Collins et al., 2003). Stratospheric-tropospheric exchange (STE) results primarily from tropopause folding along the upper extents of strong frontal boundaries and in deep low pressure systems. Stratospheric ozone is an important contributor to surface ozone concentrations in areas subject to extensive downward transport (e.g., strong high-pressure systems) or deep stratospheric intrusion events. In regional photochemical modeling applications, influences from stratospheric ozone can be mostly accommodated by extending the modeling domain into the lower stratosphere and providing boundary conditions that properly characterize stratospheric ozone levels in model layers above the local tropopause. Given the relatively confined area of a regional modeling domain, the residence time of upper-layer air is typically 1-3 days (depending on domain size and wind speeds), so boundary conditions constantly refresh the supply of stratospheric ozone on that time scale. However, this is not the case with global or hemispheric models; these models must chemically maintain or otherwise specify stratospheric ozone concentrations. STE is complicated by the highly dynamic nature of the tropopause, which varies substantially with latitude and season on top of the short-term spatio-temporal undulations induced by mid-latitude baroclinic waves. These dynamics must be adequately resolved by the grid and properly addressed by the source of meteorological data that drives the chemical transport models.

Natural stratospheric ozone chemistry is distinctly different from tropospheric chemistry involving NOx and VOC. Therefore, models that explicitly include stratospheric chemistry must address an entirely separate set of complex reactions involving specific halogens. Such mechanisms have been implemented in global chemical transport models such as AM3 (Donner et al., 2011), GEOS-Chem/UCX (Eastham et al., 2014), and GU-WRF/Chem (Karamchandani et al., 2012; Zhang et al., 2012). Such mechanisms would take time to implement and rigorously test in regional models like CAMx and CMAQ, and would likely add significantly to the computational burden. Much simpler diagnostic approaches are often implemented in global models that exploit observed correlations between lower stratospheric ozone concentration and a conserved metric from fluid dynamics called “potential vorticity” (PV) that can be used as a “tracer” for stratospheric air.

M17 describe an approach for their hemispheric CMAQ applications that use O3:PV ratios to set ozone concentrations in stratospheric layers, where PV is determined by WRF. Large variability in O3:PV has been observed (Xing et al., 2016 and references therein), ranging from 20 to over 100 ppb/PVu. Initial hemispheric CMAQ runs over the year 2006 employed a uniform O3:PV ratio of 20 ppb/PVu. The 2006 CMAQ application extended only to 100 mb (~16 km) with very coarse resolution at the top of the domain, and so the conservatively minimum value of 20 ppb/PVu was used to set stratospheric ozone only in the topmost layer to guard against over estimates of stratospheric ozone contributions in the mid- and lower troposphere. Results indicated that ozone was largely under estimated in the mid- and upper-troposphere throughout the domain. More recent hemispheric CMAQ applications achieved better overall ozone performance against global surface and aloft observations using their parameterized “dynamic” O3:PV ratio (Xing et al., 2016). Derived from global ozonesonde data and WRF-generated PV analyses over 1990-2010, the dynamic O3:PV ratio is based on a fifth-order polynomial fit that accounts for variations in latitude and season. Xing et al. (2016) derived separate parameterization for each of the top three layers of the current 44-layer CMAQ configuration that span 100 to 50 mb (16-20 km). As a result, the Xing parameterization is intimately tied to that particular layer structure, but they stress again that more layers would likely improve transport to the mid-troposphere.

A similar approach could be readily adopted for hemispheric CAMx applications, especially if initial CAMx tests employ EPA’s hemispheric WRF datasets and layer structures. Besides implementing a new O3:PV algorithm, the WRFCAMx interface program would need to be updated to extract or calculate PV fields from the raw WRF output. CMAQ applications must rely on such stratospheric ozone parameterizations because of its lack of ability to know anything about the stratosphere at the top of the domain. CAMx, however, includes an option for time- and space-varying top boundary conditions (BC), and this information could be leveraged to define an evolving stratospheric ozone profile in each grid column between the top of the model and a diagnosed level of the tropopause. The profile could be simply assigned uniformly through all stratospheric layers, or based on the ozone gradient between the model top and the layer containing the tropopause. As performed currently, WRF-resolved dynamics would determine the flux of ozone through the layer containing the tropopause. Top BCs, like the lateral and initial conditions, could be generated from pre-existing global model output since their domains extend so much higher than 50 mb. This approach would allow flexibility with any vertical layer structure.

A more complicated variant of the top BC approach involves defining the CAMx model top to follow the space- and time-variable height of the diagnosed tropopause. This is possible because the CAMx vertical layer structure can be defined arbitrarily and can vary in time and space itself. Top boundary conditions would then be extracted from existing global model output above these varying heights, and WRF-derived dynamics would determine the flux across the tropopause (model top) similarly to the case described above. This would allow removal of the need for stratospheric layers, which could then be assigned to improve resolution in the mid- and upper troposphere, and the need to make any assumptions about a lower-stratospheric ozone profile. However, this approach would require major modifications to WRFCAMx, so that it could generate a hybrid vertical grid system between WRF layers and a topopause-based top, and to the global model interface programs to extract the appropriate ozone concentrations at the undulating CAMx model top. Special cases where the tropopause is folded or not clearly defined would need to be addressed. A large amount of testing and quality assurance would be necessary for each model component.

2.3.2 Sub-Grid Cloud Mixing/Chemistry/Wet Deposition

As discussed in Section 2.2.1, deep convection is an important component of three-dimensional transport at global scales, as well as a driver of aqueous chemistry and wet removal. Additionally, sub-grid convective processes below ~100 km scale are likely a large fraction of total convection around the globe. CAMx includes the CiG, which is a relatively new framework that manages convective transport, aqueous chemistry and wet removal at sub-grid scales. Without CiG, CAMx does not account for sub-grid convective transport, and only partially includes effects of sub-grid clouds on chemistry and wet deposition. Therefore, utilizing the CiG option would be an important feature for hemispheric applications.

The current implementation of CiG has been fully tested for ozone, and partially tested for PM. However, the CiG does not include connections to the Probing Tools (source apportionment, etc.). Hemispheric CAMx applications will require that the CiG sub-model is more fully developed to include Probing Tool tracers and comprehensively tested. CiG is currently dependent on information derived by WRF using the EPA's "scale-aware" version of the Kain-Fritsch (KF) scheme. According to anecdotal information from EPA, the scale-aware KF scheme will not be further updated, and is rarely used. A scale-aware version of the Grell convection scheme is also available in WRF, and is currently used for WRF-Chem applications. Given the likelihood that the Grell scheme will continue to evolve with WRF-Chem, it would be advantageous to update the WRFCAMx interface program to use output from that scheme. Based on our current understanding, the Grell scheme outputs very similar convective flux data as KF, so we anticipate that WRFCAMx modifications would be straightforward.

2.4 Interfacing with Regional CAMx Applications

The approach to interface hemispheric CAMx output to regional applications via boundary conditions will need to address the following: (1) differing map projections/resolutions; (2) differing layer structures; and (3) source apportionment tracers. To maximize chemical consistency between global and regional simulation, the same chemistry mechanisms should be employed in both cases. This will avoid uncertainties related to mapping species between different mechanisms.

Ramboll has developed a program called BNDEXTR that supports separate, 1-way nested grid CAMx applications. BNDEXTR converts three-dimensional output from a coarse grid run to initial and boundary conditions for a fine grid run. It is designed to maximize flexibility among grid projections and resolutions: it interpolates concentration data from one projection/resolution (e.g., polar at 100 km resolution) to initial/boundary conditions on a totally different projection/resolution (e.g., Lambert at 36 km resolution), as long as the target grid fits within the larger grid. However, it assumes identical layer structures between the two grids, and transfers coarse-grid concentrations for all species directly to initial/boundary conditions with no units conversions or species mapping. It also generates time-varying boundary conditions according to the coarse grid output frequency, so if hemispheric applications output at 1 or 3-hourly intervals, boundary conditions would be derived at those frequencies. Therefore, BNDEXTR is the logical starting point for a hemispheric-regional domain interface. The program may need to be updated to accommodate different layer structures for increased flexibility.

2.4.1 Source Apportionment Updates for Hemispheric-Regional Coupling

Currently, the source apportionment tool tracks separate contributions from initial and boundary conditions, where boundary conditions can represent a single aggregated “source” contribution, or disaggregated into the 5 specific sources representing each edge (west, east, north, south, top). Boundary conditions only provide a single “source” of ozone, PM2.5 and precursors; no information is provided for source regions/categories from outside the grid. In the case of 1-way nested simulations, where each grid is run separately with its own boundary conditions, any source apportionment information derived on the coarser grid is lost when downscaling boundary conditions to the nested grid. Clearly, this process is not useful for 1-way hemispheric-regional CAMx simulations where international source apportionment is to be tracked.

A specialized program, branching from BNDEXTR, will be needed to process gridded source apportionment tracer output from a hemispheric CAMx run to a new specialized set of source apportionment boundary conditions to carry through to the regional run. It may also be necessary to include additional boundary metadata for each tracer to identify its source region and category from the hemispheric configuration.

CAMx will need to be updated to read these alternative source apportionment boundary conditions, restructure the internal source apportionment arrays to handle region/category-specific boundary conditions, and combine hemispheric source region/category information into the matrix of regional-domain source region/category tracers. For example, say CAMx/OSAT is run on the hemisphere tracking 5 source regions and 4 emission categories. Boundary conditions must be developed for that matrix of 20 regions/categories for each tracer class (ozone, NOx precursors, VOC precursors). Then say that the regional CAMx/OSAT run tracks 10 US regions with 6 emission categories. CAMx must be able to read the new file of tracer boundary conditions, add the 5 global regions to the 10 US regions for a total of 15 regions, add the 4 global categories to the 6 US categories for a total of 10 categories, and proceed with a total matrix of 150 regions/categories for each tracer class. Note that the OSAT matrix can balloon significantly depending on the combination of global and regional tracers to run. It may be possible to blend common source categories between the hemispheric and regional OSAT applications to maximize efficiency. From the example above, if the 4 global source categories are defined by the user to be equivalent to 4 US source categories (e.g., mobile, EGU, industrial, area), then the 4 global category boundary conditions would be mapped into the 4 parallel regional categories, resulting in a total matrix of 90 regions/categories for each tracer class, as opposed to 150.

3.0 CONCLUSION

This scoping study develops the foundation for a CAMx hemispheric modeling platform. Based on a literature review of current hemispheric and global modeling systems, we investigate approaches and identify technical hurdles related to configuring and running CAMx for the entire northern hemisphere. From these reviews, we have gained insights into data needs and sources, horizontal/vertical grid structures, needed technical updates, and interfacing techniques with TCEQ's regional grids. Developing a CAMx hemispheric modeling platform would provide a consistent, stream-lined global-to-regional modeling system that would simplify the TCEQ's modeling procedures and add important Probing Tools such as Source Apportionment across the northern hemisphere.

3.1 Recommended Approach for Proof-of-Concept Project

We have developed a recommended approach, and estimated cost and schedule, for a "proof-of-concept" implementation of CAMx at the hemispheric scale, including necessary software development and testing. The approach includes a list of model and pre-processing updates and improved data sources needed to better address chemical and physical processes at the global scale. Our approach is guided by information and recommendations stemming from this scoping study.

To minimize costs for the proof-of-concept application, the project would leverage the hemispheric datasets (1990-2010) and modeling approaches that have been developed previously by EPA (Mathur et al., 2017) as much as possible. These would include domain/grid specifications, meteorological fields, and global emission inventories (anthropogenic, biogenic, fires). Data developed specifically for the hemispheric project would include initial and lateral/top boundary conditions from existing global model output (especially important for stratospheric ozone), and wind-blown dust, sea salt and lightning NOx emissions generated using existing CAMx pre-processors. CAMx would be run for one year of EPA's hemispheric meteorological and emissions data, employ the most current CB6r4 chemistry mechanism, and then be evaluated against available global measurements. An initial simple application of hemispheric source apportionment would be tested, including any CAMx modifications necessary to account for the stratospheric ozone treatment. A final report would document datasets employed, CAMx modifications, application strategy, and evaluation results. The report would recommend necessary follow-on model improvements and additional hemispheric data development.

Specific details of the recommended approach are listed below; a few options are included parenthetically below certain items, which would increase the baseline cost and schedule estimates. These options could be considered for follow-on projects.

1) Domain: set according to the M17 CMAQ polar stereographic projection and horizontal resolution; include layers up through the top of WRF (50 mb) to resolve the dynamics of the upper troposphere and lower stratosphere;

2) Meteorological Data and Coupling: select a year from existing M17 hemispheric WRF datasets from 1990-2010, process through WRFCAMx; consider no layer collapsing to maximize meteorological consistency and minimize vertical diffusion of international transport plumes;

   (Options: run WRF for a year more pertinent to TCEQ projects, employ a sub-grid convection scheme that can support the CAMx CiG, evaluate WRF performance; test impacts of higher vertical resolution on the diffusion of internationally transported plumes);

3) Deep convection: use the cloud patch within the KVPATCH adjustment program to increase vertical mixing through the depth of sub-grid convective clouds, which would require extending the cloud patch to work over oceans;

   (Options: implement Probing Tools into CiG, test comprehensively, use CiG to explicitly model deep convection; extend WRFCAMx to include the multi-scale Grell convective scheme as a driver for CiG);

4) Initial conditions: derive from MOZART data, reducing model spinup to perhaps 1 month;

5) Boundary conditions: derived from MOZART data;

6) Emission inventories: use M17 hemispheric datasets derived for CMAQ/CB05TU, with any necessary species re-mapping to CB6r4;

7) Wind-blown dust (WBD) emissions: use the new CAMx WBD pre-processor;

8) Marine emissions: use the CAMx SEASALT processor with halogens needed for CB6r4;

9) Lightning emissions: use the new CAMx LNOx processor, planned for development during summer 2018;

10) Chemistry: use CB6r4 + CF/SOAP/ EQSAM;

11) Stratospheric-tropospheric ozone exchange: derive top boundary conditions from MOZART data, modify CAMx to reset ozone in stratospheric layers according to time/space-variable top boundary conditions;

   (Option: implement the uniform or dynamic O3:PV ratio approach from CMAQ);

12) CAMx application: run for the selected year and conduct a basic preliminary evaluation against global surface observation data;

   (Option: run CAMx for 2006 and conduct a more thorough evaluation against INTEX-B datasets similar to the M17 CMAQ approach);

13) Demonstrate a simple OSAT case: update CAMx to account for source apportionment boundary conditions and stratospheric ozone; run a simple case for a short period of 1 month;

14) Reporting: document datasets employed, CAMx modifications, application strategy, and evaluation results; recommend necessary follow-on model improvements and additional hemispheric data development.

The estimated cost to complete the baseline proof-of-concept project listed above, not including the optional components, ranges $115,000 to $125,000. The estimated schedule for completion is 8-9 months. Including optional elements listed above may increase the overall budget by as much as $150,000 and extend the project schedule by up to 9 months, depending on the options selected.

4.0 REFERENCES

Appel, K., et al., 2017. Description and evaluation of the Community Multiscale Air Quality (CMAQ) modeling system version 5.1. *Geoscientific Model Development*, 10(4), 1703-1732. doi: 10.5194/gmd-10-1703-2017.

Astitha et al., 2012. Parameterization of dust emissions in the global atmospheric chemistry-climate model EMAC: impact of nudging and soil properties. *Atmos. Chem. Phys.*, 12(22):11057-11083, doi:10.5194/acp-12-11057-2012.

Bey, I., D.J. Jacob, R.M. Yantosca, J.A. Logan, B.D. Field, A.M. Fiore, Q. Li, H.Y. Liu, L.J. Mickley, M.G. Schultz, 2001. Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation. *J. Geophys. Res. Atmos.*, 106(D19), 23073–23095, 2001.

Collins, W. J., Derwent, R. G., Garnier, B., Johnson, C. E., Sanderson, M. G., and Stevenson, D. S., 2003. Effect of stratosphere troposphere exchange on the future tropospheric ozone trend. *J. Geophys. Res.*, 108, 8528, doi:10.1029/2002JD002617, 2003.

Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., Van Dingenen, R., Granier, C., 2016. Forty years of improvements in European air quality: regional policy-industry interactions with global impacts. *Atmos. Chem. Phys*., 16, 3825-3841, https://doi.org/10.5194/acp-16-3825-2016.

Donner, L.J., Wyman, B.L., Hemler, R.S., Horowitz, L.W., Ming, Y., Zhao, M., Golaz, J.C., Ginoux, P., Lin, S.J., Schwarzkopf, M.D., Austin, J., Alaka, G., Cooke, W.F., Delworth, T.L., Freidenreich, S.M., Gordon, C.T., Griffies, S.M., Held, I.M., Hurlin, W.J., Klein, S.A., Knutson, T.R., Langenhorst, A.R., Lee, H.C., Lin, Y., Magi, B.I., Malyshev, S.L., Milly, P.C.D., Naik, V., Nath, M.J., Pincus, R., Ploshay, J.J., Ramaswamy, V., Seman, C.J., Shevliakova, E., Sirutis, J.J., Stern, W.F., Stouffer, R.J., Wilson, R.J., Winton, M., Wittenberg, A.T., Zeng, F., 2011. The Dynamical Core, Physical Parameterizations, and Basic Simulation Characteristics of the Atmospheric Component AM3 of the GFDL Global Coupled Model CM3. *J. Climate*, 24, 3484–3519, http://dx.doi.org/10.1175/2011JCLI3955.1.

Eastham, S.D., D.K. Weisenstein, S.R.H. Barrett, 2014. Development and evaluation of the unified tropospheric–stratospheric chemistry extension (UCX) for the global chemistry-transport model GEOS-Chem. *Atmos. Environ*., 89, 52-63, https://doi.org/10.1016/j.atmosenv.2014.02.001.

Eastham, S.D. and D.J. Jacob, 2017. Limits on the ability of global Eulerian models to resolve intercontinental transport of chemical plumes. *Atmos. Chem. Phys.*, 17(4), 2543–2553, doi:10.5194/acp-17-2543-2017, 2017.

Emmons, L.K., Walters, S., Hess, P.G., Lamarque, J.F., Pfister, G.G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S.L., Kloster, S., 2010. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). *Geosci. Model Dev.*, 3, 43-67, doi:10.5194/gmd-3-43-2010.

Foroutan, H., Young, J., Napelenok, S., Ran, L., Appel, K.W., Gilliam, R.C., Pleim, J.E., 2017. Development and evaluation of a physics-based windblown dust emission scheme implemented in the CMAQ modeling system. *J. Advances in Modeling Earth Systems,* 9, 585-608, doi: 10.1002/2016MS000823.

Granier, C., Bessagnet, B., Bond, T.C., Klimont, Z., Riahi, K., 2011. Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980-2010 period. *Climatic Change* 109 (1), 163-190, doi:10.1007/s10584-011-0154-1.

Harvard University, 2018. GEOS-Chem Model website. http://acmg.seas.harvard.edu/geos.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., Zhang, Q., 2018. Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS). *Geosci. Model Dev*., 11, 369-408, https://doi.org/10.5194/gmd-11-369-2018.

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., Li, M, 2015. HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution. *Atmos. Chem. Phys*., 15, 11411-11432, https://doi.org/10.5194/acp-15-11411-2015.

Karamchandani, P., Zhang, Y., Chen, S.-Y., Balmori-Bronson, R., 2012. Development of an extended chemical mechanism for global–through–urban applications. *Atmos. Poll. Res.* 3, 1-24, doi: 10.5094/APR.2011.047.

Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., Schöpp, W., 2017. Global anthropogenic emissions of particulate matter including black carbon. *Atmos. Chem. Phys*., 17, 8681-8723, https://doi.org/10.5194/acp-17-8681-2017.

Klingmueller et al, 2017. Revised mineral dust emissions in the atmospheric chemistry-climate model EMAC (based on MESSy 2.52). *Geosci. Model Dev. Discuss.*, https://doi.org/10.5194/gmd-2017-160.

Lamarque, J.F., Bond, T.C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M.G., Shindell, D., Smith, S.J., Stehfest, E., Van Aardenne, J., Cooper, O.R., Kainuma, M., Mahowald, N., McConnell, J.R., Naik, V., Riahi, K., van Vuuren, D.P., 2010. Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmos. Chem. Phys.* 10, 7017-7039, doi: 10.5194/acp-10-7017-2010.

Lelieveld, J. and Dentener, F. J., 2000. What controls tropospheric ozone? *J. Geophys. Res.*, 105, 3531–3551.

Marsh, D.R. M.J. Mills, D.E. Kinnison, J.-F. Lamarque, N. Calvo, L.M. Polvani, 2013. Climate Change from 1850 to 2005 Simulated in CESM1(WACCM). *J. Climate*, https://doi.org/10.1175/JCLI-D-12-00558.1.

Mathur, R., J. Xing, R. Gilliam, G. Sarwar, C. Hogrefe, J. Pleim, G. Pouliot, S. Roselle, T.L. Spero, D.C. Wong, J. Young, 2017. Extending the Community Multiscale Air Quality (CMAQ) modeling system to hemispheric scales: overview of process considerations and initial applications. *Atmos. Chem. Phys.*, 17, 12449–12474.

NCAR, 2018. Global Chemistry Modeling web site: https://www2.acom.ucar.edu/gcm/mozart.

Ramboll, 2018. User's Guide: the Comprehensive Air quality Model with extensions, Version 6.50. Available at http://www.camx.com/files/camxusersguide_v6-50.pdf.

Sarwar, G., Godowitch, J., Henderson, B.H., Fahey, K., Pouliot, G., Hutzell, W.T., Mathur, R., Kang, D., Goliff, W.S., Stockwell, W.R., 2013. A comparison of atmospheric composition using the Carbon Bond and Regional Atmospheric Chemistry Mechanisms. *Atmos. Chem. Phys.*, 13, 9695–9712. https://doi.org/10.5194/acp-13-9695-2013.

Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Dudha, M.G., Huang, X., Wang, W., Powers, Y., 2008. A description of the advanced research WRF Ver. 30. NCAR Technical Note (p. 113). NCAR/TN-475.

Stohl, A., Aamaas, B., Amann, M., Baker, L. H., Bellouin, N., Berntsen, T. K., Boucher, O., Cherian, R., Collins, W., Daskalakis, N., Dusinska, M., Eckhardt, S., Fuglestvedt, J. S., Harju, M., Heyes, C., Hodnebrog, Ø., Hao, J., Im, U., Kanakidou, M., Klimont, Z., Kupiainen, K., Law, K. S., Lund, M. T., Maas, R., MacIntosh, C. R., Myhre, G., Myriokefalitakis, S., Olivié, D., Quaas, J., Quennehen, B., Raut, J.-C., Rumbold, S. T., Samset, B. H., Schulz, M., Seland, Ø., Shine, K. P., Skeie, R. B., Wang, S., Yttri, K. E., Zhu, T., 2015. Evaluating the climate and air quality impacts of short-lived pollutants. *Atmos. Chem. Phys*., 15, 10529-10566, https://doi.org/10.5194/acp-15-10529-2015.

Xing, J., Mathur, R., Pleim, J., Hogrefe, C., Gan, C.-M., Wong, D.C., Wei, C., Gilliam, R., Pouliot, G., 2015. Observations and modeling of air quality trends over 1990–2010 across the Northern Hemisphere: China, the United States and Europe. *Atmos. Chem. Phys.*, 15, 2723–2747, https://doi.org/10.5194/acp-15-2723-2015.

Xing, J., R. Mathur, J. Pleim, C. Hogrefe, J. Wang, C.-M. Gan, G. Sarwar, D.C. Wong, S. McKeen, 2016. Representing the effects of stratosphere–troposphere exchange on 3-D O3 distributions in chemistry transport models using a potential vorticity-based parameterization. *Atmos. Chem. Phys.*, 16, 10865–10877, doi:10.5194/acp-16-10865-2016.

Zhang, Y., Karamchandani, P., Glotfelty, T., Streets, D.G., Grell, G., Nenes, A., Yu, F., Bennartz, R., 2012. Development and initial application of the global-through-urban weather research and forecasting model with chemistry (GU-WRF/Chem). *J. Geophys. Res., Atmospheres* 117, D20206, doi: 10.1029/2012jd017966.

Zhuang, J., D.J. Jacob, S.D. Eastham, 2017. The importance of vertical resolution in the free troposphere for modeling intercontinental plumes. *Atmos. Chem. Phys. Discuss.*, https://doi.org/10.5194/acp-2017-1124.