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Improving the Characterization of Natural Emissions in CMAQ

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Introduction

The air quality of any given location is the result of complex interactions between meteorology and emissions through dynamical, physical, chemical, and photochemical processes. Of the many sources of air pollution, naturally produced emissions such as nitric oxide (NO) from soil[1] and lightning[2], biogenic volatile organic compounds (BVOCs) [3] from vegetation, and sulfur dioxide (SO_2) and carbon dioxide (CO_2) from volcanic eruptions[4] constitute a nonnegligible and relatively stable portion of the total emissions of these pollutants. Despite the decreasing trend of anthropogenic emissions in the United States (U.S.), surface-level ozone (O_3) concentrations have not subsequently decreased in many regions including the Mountain West and the Pacific Coast[5]. This seemingly contradictory result was further revealed by the lockdowns instituted to control the coronavirus disease 2019 (COVID-19) pandemic. During that period in the Spring of 2020, surface O_3 levels increased by up to 50% in some locations despite large and widespread reductions in loading from fossil fuel NO emissions associated with vehicles[6]. In addition to the nonlinear atmospheric chemistry under decreasing anthropogenic emissions, changes in the relative contribution of natural emissions to the total emissions budget in the atmosphere become increasingly important[7]. For example, the oceanic emissions of iodine have tripled since 1950, driven by anthropogenic O_3 pollution and rising temperatures, as registered by Arctic and Alpine ice cores and tree ring measurements[8–10]. To quantify the complex impacts of natural emissions on air quality, chemical transport models must include realistic characterizations of the source strengths, spatial and temporal distributions of the emissions, and evolution of the emitted species from natural sources due to climate change. Recent updates to the U.S. Environmental Protection Agency's (EPA's) Community Multiscale Air Quality (CMAQ) modeling system have included improvements to the emission and chemistry of natural emissions. In this paper, we summarize the updates to

lightning NO_x (LNO_x), BVOCs, soil NOx (SNO_x), and halogen emissions in CMAQ and their impacts on model predicted surface O_3.

Lightning NO_x Parameterization

Previous studies have shown that about 10–15% of the total global NO_x emissions originates from LNO_x, which accounts for 35–50% of global free-tropospheric O_3[11,12]. The contribution of LNO_x to surface O_3 can be significant at high elevation locations including the U.S. Intermountain West, where LNO_x may contribute 3–4 ppbv to the annual mean 8-hr maximum O_3 and up to 18 ppbv during episodic events[2,13]. In addition, lightning activity and the associated distribution of LNO_x exhibit strong geographic and temporal variations[7]. Different options to represent LNO_x production have been implemented beginning with CMAQv5.2[14–15]. For retrospective (i.e., historical) simulations, hourly lightning flashes from ground-based lightning observation networks are used, when observed lightning data are unavailable (such as in future climate studies and air quality forecasts), a LNO_x parameterization is available that use climatological relationships associated with convective precipitation (CP) and observed lightning flashes[14].

The initial implementation of the climatological LNO_x parameterization in CMAQ was formulated using available modeled CP data from the Weather Research and Forecasting (WRF) model versions (3.4 to 3.8) and observed lightning flashes from the National Lightning Detection Network (NLDN) from 2002 to 2014[14]. Leveraging the EPA's Air QUAlity TimE Series (EQUATES) project in which WRFv4.1.1 is consistently used to produce the meteorological fields from 2002 to 2018, the LNO_x parameterization scheme was reformulated to consider the longer time period and unified CP prediction from a single version of WRF. In addition, the updated WRF dataset presented an opportunity to assess the reliability of the methodology over time and space. For that purpose, the parameters are formulated with the following time periods: 2002–2014 (the same period used to develop the original parameterization), 2002–2009, and 2010–2018 (for assessment of consistency over non-overlapping time periods). The climatological regression relationships between monthly CP and lightning flashes for all parameterizations are comparable across geographic regions, indicating that the original methodology is reliable and consistent. Figure 1a displays the total monthly LNO_x emissions generated by these different parameter sets and hourly NLDN lightning flashes when they are applied to the 2016 annual CMAQ simulations. Examples of the spatial distributions of LNO_x emissions generated by hourly NLDN lightning flashes and one of the parameterizations in July 2016 are shown in Figure 1b and Figure 1c, respectively. The LNO_x emissions from these parameterizations closely follow the NLDN case for the monthly variations, but they are 10–20% lower than those generated by directly using the NLDN data during the warm months. These reduced LNO_x emission rates had a minor impact on surface O_3 concentrations[15].

Biogenic VOCs and Soil NO_x

Highly reactive BVOCs such as isoprene are abundant[16] and their rapid oxidation results in O_3 formation. The Biogenic Emission Inventory System (BEIS) model has been the only option for simulating inline BVOC with CMAQ since its implementation in 2002[17].

BEIS calculates BVOC as a function of simulated meteorology and vegetation data, including the Biogenic Emission Landuse Database (BELD). Since BELD is currently only available for domains within North America, using BEIS for other geographic domains requires additional user development. To update and expand the inline BVOC capability in CMAQ, a second BVOC option (the Model for Emissions of Aerosols and Gasses from Nature (MEGAN) version 3.1[18]) will be available in future releases of CMAQ. MEGAN calculates BVOC emissions as the product of emission activity, a function of meteorological conditions and plant stress factors, and time-independent averages of emission rates that are prepared using the MEGAN preprocessor.

July is the month with the most emitted isoprene over the Northern Hemisphere [19] and a time when we expect to see large sensitivities to different BVOC model options. Results from a July 2016 simulation using a configuration[20] for the U.S. illustrate differences in isoprene from the CMAQ implementations of MEGAN and BEIS (Figure 2a,b). The domain total isoprene emissions for this month are 4.63 Tg for MEGAN and 4.26 Tg for BEIS. When the drought stress setting for MEGAN is enabled, the emissions of isoprene decrease roughly 30% to 3.27 Tg. Reduction of isoprene emissions due to MEGAN's parameterization of drought stress varies from 7%[16] to 21%[21] for global emissions, and up to 68%[3] for regional emissions. Reducing BVOC emissions due to drought sensitivity improves model skill for air quality metrics in the CMAQ and MEGAN configuration used in Figure 2.

Soil NO from agricultural and non-agricultural sources is included as biogenic emissions in both BEIS and MEGAN. Both BVOC models use a soil NO calculation based on Yienger and Levy (YL)[1], but the MEGAN implementation produces less soil NO due to different emissions factors (Figure 2c,d). MEGAN 3.1 also introduces the Berkley Dalhousie Soil NO Parameterization (BDSNP) [22], which produces significantly more soil NO than either YL option (not shown).

The effect of BVOC modeling choice on O_3 is ultimately small (Figure 3), reducing monthly-mean bias by up to 2 ppb relative to BEIS in the midwestern U.S. The reduction in high O_3 bias coincides with the decreased soil NO emissions in the MEGAN implementation. In the western U.S., the O_3 underprediction in the BEIS simulation is worsened in the MEGAN implementation. These comparisons highlight the challenges/uncertainties in representing biogenic emissions in current air quality models. Having two BVOC models in CMAQ enables users to pick the suitable option for their specific application and facilitates quantification of the uncertainties associated with biogenic emissions.

Halogen Emissions and Chemistry

Bromine and iodine (hereafter "halogen") compounds emitted from the oceans contribute to atmospheric chemistry and reduce O_3 concentrations over seawater, as well as in some coastal and inland areas[23]. Marine halogen emissions in CMAQv5.3.2 include both organic and inorganic compounds. The organic halogen compounds, collectively known as halocarbons, are produced by phytoplankton and macroalgae[24]. Specifically, emissions

of nine halocarbon species are included in CMAQv5.3.2 and are estimated using monthly climatological chlorophyll-a concentrations retrieved from the Moderate Resolution Imaging Spectroradiometer (MODIS) satellite. The emissions rates of inorganic halogen compounds are a function of sea surface temperature and 10-meter wind speed[25] and included in CMAQv5.3.2.

CMAQv5.3.2 contains a detailed treatment of bromine and iodine reactions[23] which are further updated for the next version of CMAQ model. Three sets of annual simulations were completed over the Northern Hemisphere for 2016 by using: (1) updated halogen emissions/chemistry (2) existing halogen emissions/chemistry in CMAQv5.3.2, and (3) no halogen emissions/chemistry. The halogen updates increase seasonal mean O_3 over seawater and land areas compared to the existing emissions/chemistry (Figure 4a–d). The impacts are higher in winter months than those in summer months. Existing halogen emissions and chemistry in CMAQv5.3.2 reduces annual mean surface O_3 by ~21% compared to the simulation without halogens while the updated halogen emissions and chemistry reduces surface O_3 by ~16% over seawater.

Summary

EPA’s premier modeling system (CMAQ) helps shape up and/or analyze policy scenarios to support air quality management decisions. Representation of various natural emissions in CMAQ are continually being improved, and their impact on surface air quality are being assessed. Differing levels of improvement of surface O_3 retrospective predictions are achieved from the updates of different natural emissions with more options available for a variety of applications. With the decreasing trend of anthropogenic emissions and the increasing trend of global warming, efforts to improve the characterization of natural emissions in air quality models will be increasingly desirable.

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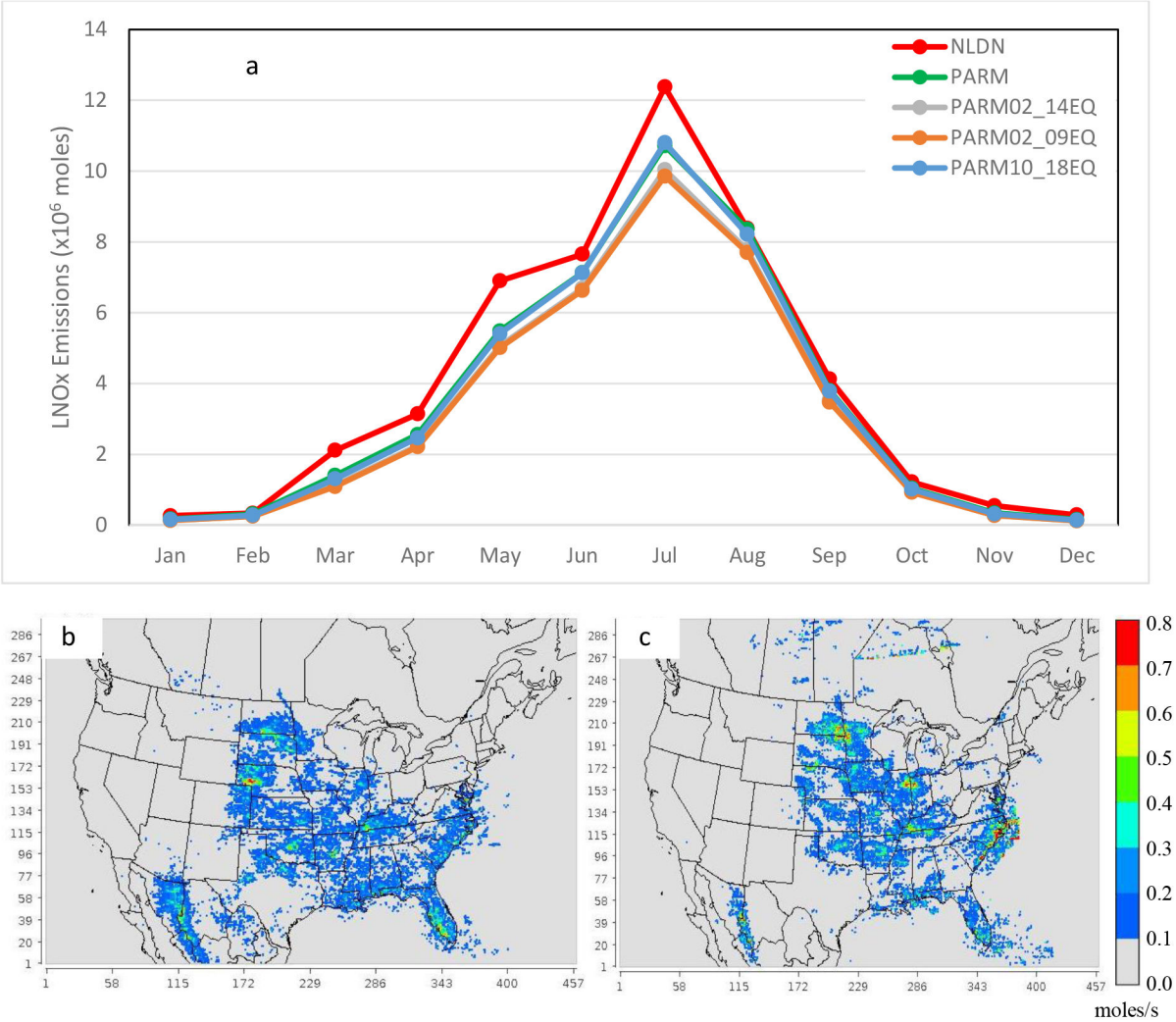

Figure 1.

(a) The monthly LNO_x emissions over the contiguous U.S. from 2016 annual CMAQ simulations using NLDN (hourly lightning flashes), PARM (the existing parameters), PARM02_14EQ (parameters formulated using WRF4.1.1 and input data period from 2002–2014), PARM02_09EQ (input data from 2002–2009), and PARM10_18EQ (input data from 2010–2018). (b) The monthly mean hourly LNO_x emissions using NLDN in July 2016. (c) The monthly mean hourly LNO_x emissions using PARM in July 2016

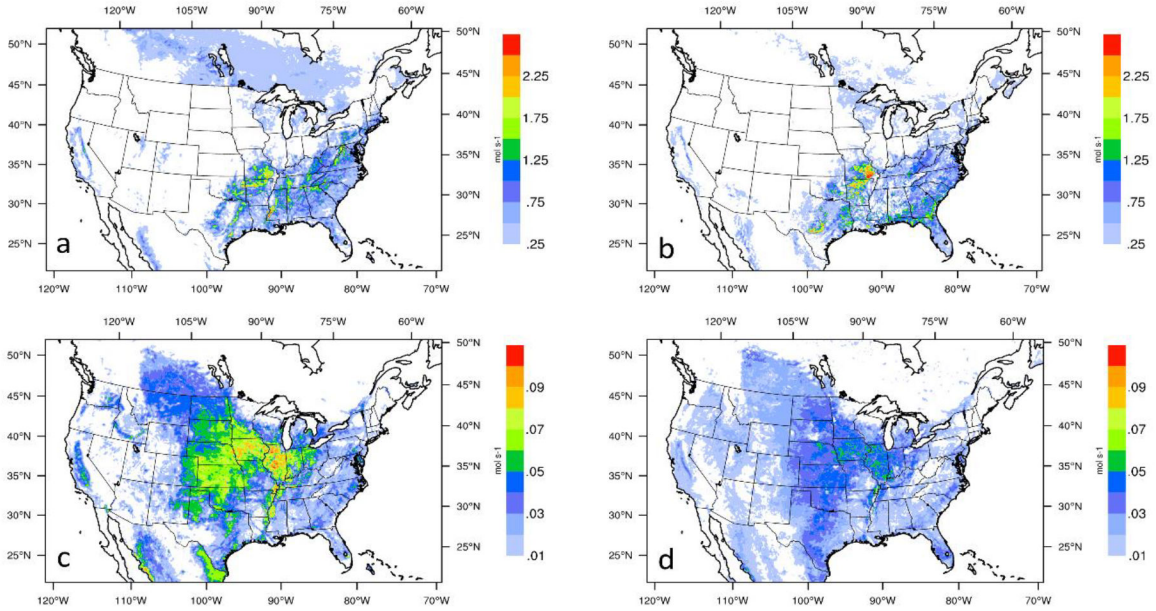

Figure 2.

Average July 2016 isoprene emissions rates (moles/s) for (a) CMAQ-BEIS and (b) CMAQ-MEGAN with drought stress option enabled. Average July 2016 soil NO emission rates (moles/s) for (c) CMAQ-BEIS and (d) CMAQ-MEGAN.

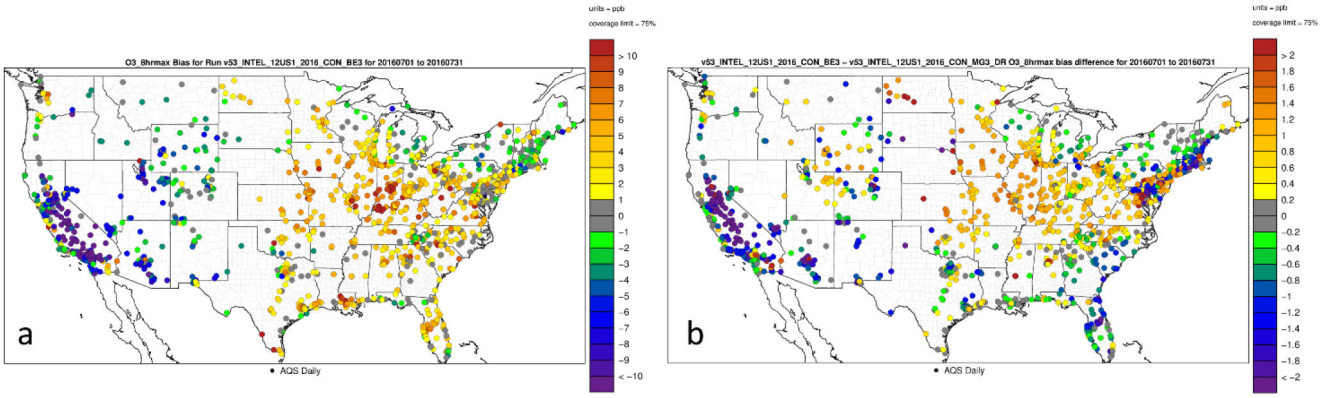

Figure 3.

(a) July 2016 daily 8-hr maximum O_3 bias for CMAQ-BEIS. (b) Difference in July 2016 daily 8-hr maximum O_3 absolute bias between CMAQ-BEIS and CMAQ-MEGAN with drought stress option enabled, where positive values indicate larger bias when using CMAQ-BEIS. Observations for bias values are from EPA's Air Quality System (AQS).

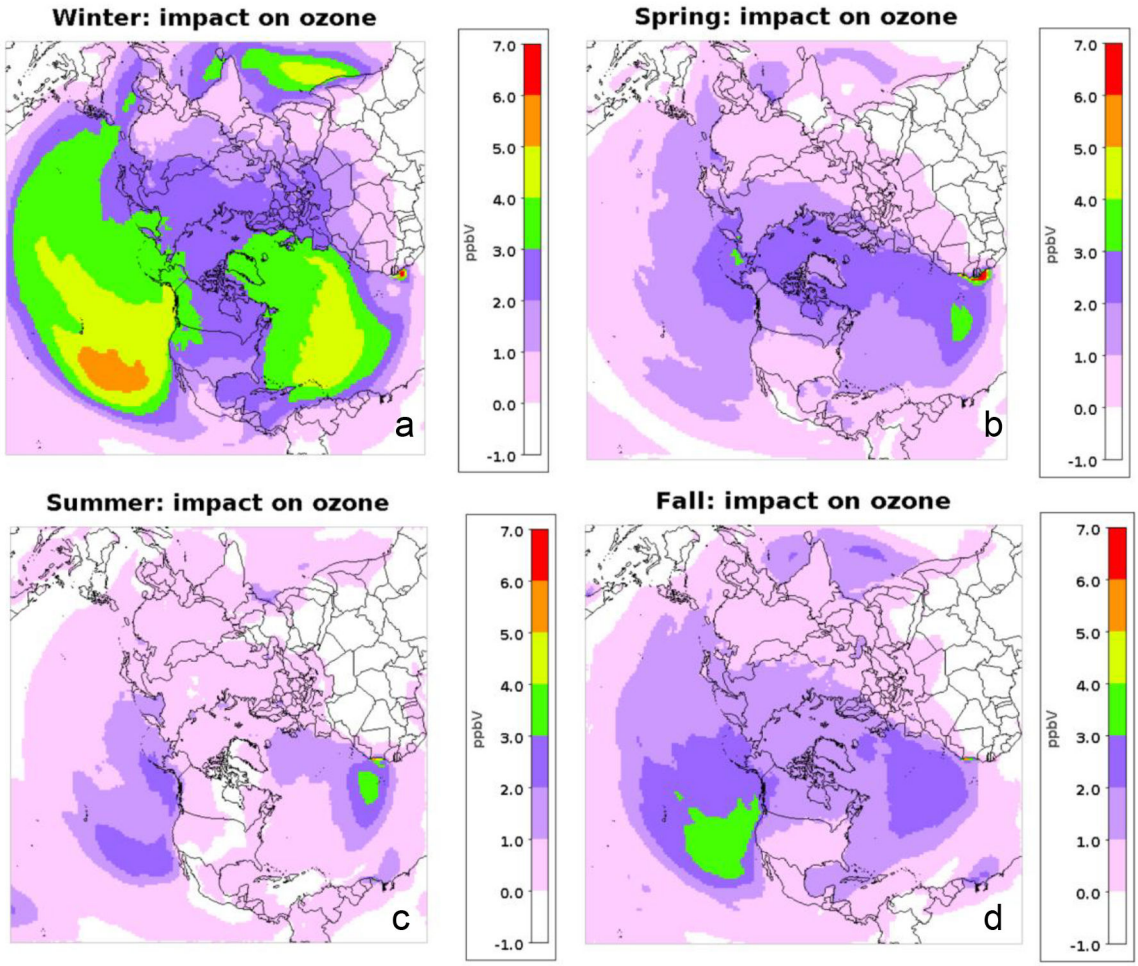

Figure 4.

Seasonal mean impact of updated halogen emissions and chemistry compared to the existing halogen emissions and chemistry in CMAQv5.3.2 on O_3 over the Northern Hemisphere (a) Winter, (b) Spring, (c) Summer, (d) Fall.