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Article

Gas-Phase Water-Soluble Organic Carbon: CMAQ Model Evaluation in Baltimore County

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Ellie N. Smith, Kirk R. Baker, Marwa M.H. El-Sayed, Christopher J. Hennigan, Simon Rosanka, and Annmarie G. Carlton*

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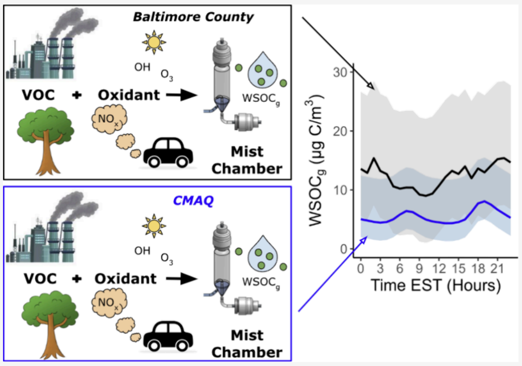
ABSTRACT: Prediction of gas-phase water-soluble organic carbon ($WSOC_g$), a precursor for secondary organic aerosol formed through processing in atmospheric waters (aqSOA), has not yet been evaluated in models. We pair the $WSOC_g$ predictions from the U.S. EPA's Community Multiscale Air Quality (CMAQ) model with continuous mist chamber measurements during February−March 2015 and August 2016 in Baltimore County, MD. We simulate mist chamber collection of $WSOC_g$ from CMAQ's atmosphere with application of compound-specific collection efficiencies as a function of Henry's law. CMAQ predictions of $WSOC_g$ mass concentrations are highest in August, while measurements are highest during February−March. CMAQ does not replicate the average diurnal pattern of the measured $WSOC_g$ in any month. The CMAQ prediction of directly emitted VOCs that oxidize to form $WSOC_g$ is more reasonable, and the model skill for nitrogen dioxide (NO_2) and ozone (O_3) is relatively excellent in comparison ($R^2 = 0.5$ and $R^2 = 0.6$, respectively; $p \approx 0$). These findings suggest that representation of organic gases and their chemistry in this CMAQ simulation is sufficient to accurately predict the criteria pollutants NO_2 and O_3, but not necessarily the chemical transformations that produce $WSOC_g$, an important precursor for aqSOA.



KEYWORDS: *air quality, CMAQ, water-soluble organic gases, OVOCs, mist chambers*

INTRODUCTION

Oxidation of volatile organic compounds (VOCs) in the atmosphere affects ozone (O_3) formation and contributes to ambient fine particulate matter ($PM_{2.5}$) through formation of semivolatile and water-soluble gases that partition to the condensed phase and form secondary organic aerosol (SOA).[1−11] Accurate description of atmospheric chemical processing is vital to understanding the fate and transport of trace species and provides insight into the impacts of controllable emissions on air quality.[12,13] The chemistry in EPA's Community Multiscale Air Quality (CMAQ) model is routinely improved as the model is widely employed for forecasting and in regulatory and research applications.[14] CMAQ is extensively evaluated for the predictive skill of criteria pollutants such as O_3, NO_2, and $PM_{2.5}$ at routine surface monitoring sites[14−18] and with satellite retrievals.[19,20] More chemically detailed model evaluation is usually limited to intensive observation periods (IOPs).[21−24]

CMAQ predicts aqueous chemical pathways that form SOA (aqSOA) are a major contributor to ambient $PM_{2.5}$ in the eastern United States,[12] especially the southeast.[25−27] CMAQ model development with a focus on aqueous chemistry improves measurement-model agreement for surface and aloft particulate matter in the United States.[25,26,28−30] CMAQ also predicts that ambient gas-phase water-soluble organic carbon ($WSOC_g$), a precursor for aqSOA, is a ubiquitous reservoir of reactive carbon throughout the atmosphere above the contiguous United States (CONUS).[25,27,31−33] Ambient surface measurements of $WSOC_g$ with mist chambers in rural, urban, marine, and Arctic environments are qualitatively consistent with estimates that suggest that $WSOC_g$ is an abundant trace species in diverse locations.[34,35] The measured $WSOC_g$ in rural and suburban surface locations of the United States, in addition to Beijing, also demonstrates positive correlations with air quality metrics, such as PM, O_3, and NO_x.[27,35−38] There is presently little quantitative understanding of the CMAQ predictive skill for the amount and statistical correlations of $WSOC_g$ with other trace species. Effective management of air quality can be

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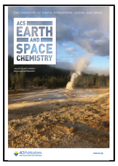

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hindered by uncertainty in precursor gases, which contributes to model bias for SOA and $PM_{2.5}$ mass predictions.[39] Prediction of $WSOC_g$, an important SOA and $PM_{2.5}$ precursor,[1,40] is poorly constrained, and this is a key knowledge gap.[41]

Gas-phase organic compounds in the chemical mechanisms employed in CMAQ and other atmospheric models are aggregated and described with surrogate species to facilitate computational speed while retaining the predictive skill for criteria pollutants, primarily ozone.[14,42−46] For example, in the carbon-bond gas-phase chemical mechanism often employed in CMAQ simulations, "PAR" is a surrogate species for paraffinic carbon. Emissions of C−C single bond species such as ethane, propane, and butane are mapped to PAR. Atmospheric processes (e.g., oxidative chemistry, wet and dry deposition) are simulated for the lumped PAR species that is assigned the chemical properties (e.g., Henry's law value and reaction rate constant with the hydroxyl radical ($^\bullet$OH) of ethane. Lumped VOC inventories can be evaluated by proxy. A common metric for the predictive skill of overall VOC inventory reactivity is the comparison of measured VOC loss due to the hydroxyl radical ($^\bullet$OH) reaction with the predicted sum of individual model VOCs that react with $^\bullet$OH: $\Sigma(\mathrm{VOC}_i \times k_{\mathrm{OH\text{-}VOCi}})$.[47] Individual VOCs measured can also be mapped to the corresponding surrogates and compared. Such evaluation of CMAQ simulations at surface sites in the northeast finds that the carbon bond 4 (CB4) chemical mechanism predicts ozone well but overpredicts all lumped VOCs.[48] CMAQ evaluation in both urban and forested areas suggests that gas-phase mechanism development to improve the representation of primary and oxygenated VOCs is needed to improve model effectiveness.[18,36,48,49]

In this work, we evaluate CMAQ in a semiquantitative way by comparing hourly average bulk values of measured and predicted $WSOC_g$ mass concentrations and observed versus predicted correlations of $WSOC_g$ with meteorological variables and air quality metrics. We compare total organic carbon (TOC) collected via the mist chamber (MC)[50] during three months of continuous ambient sampling at the University of Maryland, Baltimore County (UMBC),[51] a location predicted and observed to have an abundance of $WSOC_g$ and aqSOA.[12,25] We also evaluate the model at a nearby Photochemical Assessment Monitoring Station (PAMS) for criteria gases and chemically characterized VOCs that are $WSOC_g$ precursors. We pair MC and PAMS samples in space and time of day with CMAQ predictions. We sample the model's atmosphere subject to MC collection efficiencies (CEs) as a function of Henry's law (K_H) solubility as described below. We assess the overall solubility of CMAQ's organic gas inventory, analogous to reactivity evaluation techniques.[36,52−54]

METHODS

Measurements. Continuous ambient measurements of $WSOC_g$ via MC sampling in 2015 and 2016 (Table 1) took place at UMBC (39.2546 °N, −76.7139 °W), a suburban site in Baltimore, MD that are described in detail elsewhere.[50,51] Briefly, ambient air is passed through a 47 mm Teflon filter at 28 L/min to remove ambient particles before introduction into a wet scrubbing MC. The sampling cycle is a total of 14 min, with 5 min of active sampling, followed by 9 min of TOC sample analysis and MC rinsing to prepare for the next sample.[55,56] Due to evaporative loss, the final volume of sample with the dissolved gases is determined to be 7.78 ± 0.17 mL.[50] The collected sample is analyzed for organic carbon content with a TOC instrument (Model 900 Turbo, GE Analytical). Uncertainty is assumed to be 10%, but for compounds with a Henry's law value (K_H) < 1.4×10^2 M atm^{-1} and above 1.0×10^3 M atm^{-1}, the approximation is more uncertain.[50]

Table 1. Measurement Data Available at Each Location over the Specified Time Period

location	measured parameter	date	number of measurement−model pairs
UMBC	$WSOC_g$	02/04 to 03/24 2015	580
		07/31 to 08/28 2016	489
Essex County PAMS site	NO_2, O_3, temperature	2016	8732
	VOCs	07/31 to 08/28 2016	2043

PAMS measures criteria gases and VOCs.[57] VOCs are measured during the summer (Table 1) as part of the EPA's nonmethane organic carbon (NMOC) program. The PAMS site closest to UMBC is the Essex site (AQS Site Code 240053001, PQAO Maryland Department of the Environment, 39.310839 °N, −76.474487 °W), located in a neighboring CMAQ grid cell. VOCs are collected only during summer in pressurized canisters, analyzed via gas chromatography with a flame ionization detector (GC-FID), and measured with auto gas chromatography (GC) collection (PE Clarus 500 dual col).[58,59] All measurements are averaged on an hourly basis to match the time resolution of CMAQ.

CMAQ Simulation. The entire year of 2016 is modeled with CMAQ version 5.2.1 (doi: 10.5281/zenodo.1212601).[60] The chosen simulation is evaluated extensively for air quality and meteorology performance.[61,62] CMAQ is applied for a domain covering the CONUS using 12 km sized horizontal grid cells. The vertical atmosphere is resolved with 35 layers extending from the surface to the tropopause with finer resolution in the planetary boundary layer to better capture the diurnal variation in mixing height. The Carbon Bond 6 revision 3 gas-phase chemical mechanism,[63] ISORROPIA II inorganic aerosol thermodynamics,[64] aqueous phase cloud chemistry,[28] and wet and dry depositional processes are included in the simulation. Primary organic aerosol is treated as nonvolatile. SOA formation is described by semivolatile partitioning of lumped gas-phase groups, which include benzene, toluene, xylenes, naphthalene, monoterpenes, isoprene, and sesquiterpenes, and particle-phase oligomerization.[65] AqSOA is formed from glyoxal, glyoxal analogues (methylglyoxal and glycolaldehyde), and isoprene epoxydiols.[60,66] Biogenic emissions are estimated using the Biogenic Emission Inventory System (BEIS) version 3.6.1.[67] Anthropogenic emissions are based on the 2016 emissions modeling platform developed by the U.S. EPA.[68] Lateral boundary inflow is extracted from a coarser scale model simulation that covered the northern hemisphere.[69] Meteorological inputs from the Weather Research and Forecasting (WRF) model version 3.8[70] are used to develop biogenic emissions and drive atmospheric dynamics using the same domain and projection specifications as the CMAQ model simulation.

CMAQ Evaluation. We evaluate the CMAQ predictions of criteria gases and $WSOC_g$ by pairing simulations with measurements in space and time of day. Measurements are

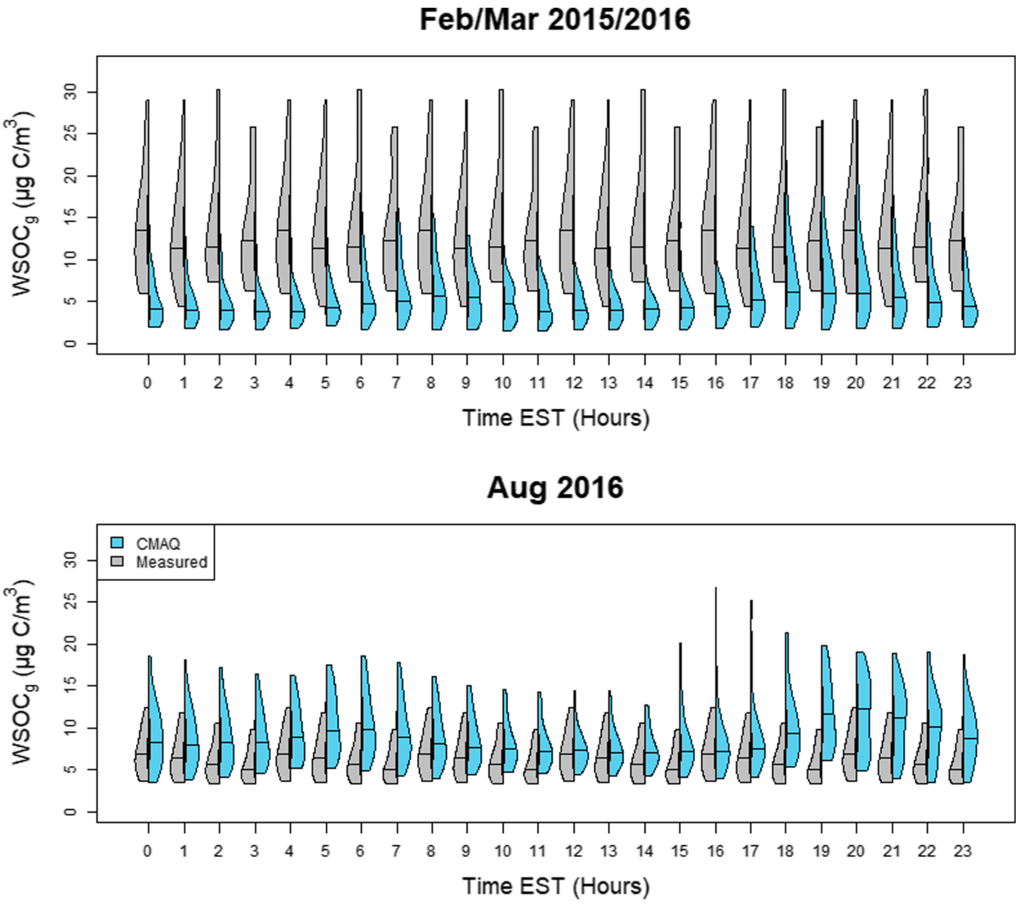

Figure 1. Average diurnal profile of MC-collected $WSOC_g$ at UMBC for (top) measurements in Feb/Mar 2015 and CMAQ in Feb/Mar 2016 and (bottom) measurements and CMAQ in Aug 2016 with simulated MC sampling in blue and measurements in gray. Time is local standard time.

aggregated to hourly values, and diurnal profiles are compared with CMAQ predictions. We evaluate the model for two separate time categories, February 4 to March 28 (2015 for measurements, 2016 for the model), and July 31 to August 28, 2016, for both measurements and model. CMAQ model time is transformed to the local standard time. Model-predicted gas-phase mixing ratios (ppb_V) are converted with the ideal gas law to mass concentrations ($\mu gC\ m^{-3}$) using WRF-predicted temperature and pressure to facilitate direct comparison with $WSOC_g$ measurements (eq 1 and eq S1). We calculate methylglyoxal equivalents ($MGLY_{equiv}$), analogous to propylene equivalents,[54] to normalize the entire gas-phase organic inventory for solubility. All organic gas concentrations are multiplied by the Henry's law constant of individual organic gas i, $K_{H\text{-}i}$ over methylglyoxal's K_H ($[VOC] \times (K_{H\text{-}i}/K_{H\text{-MGLY}})$, where $K_{H\text{-MGLY}} = 3.2 \times 10^4$ M atm^{-1}), to normalize by solubility for a chemical inventory that changes with space and time. We evaluate the seasonal and diurnal CMAQ predictions of $WSOC_g$ by simulating the MC sampling of CMAQ's atmosphere. We account for the CE of each individual gas-phase organic compound in the model (Table S1). CE accounts for compound solubility (i.e., Henry's law), sampler flow rate, and volume, in addition to evaporative loss that we assume to be constant over the sampling period (eq 2 and eq S2).[35,50]

$$c_i \times \frac{P}{RT} \times \frac{\#\text{mol C}}{\text{mol } c_i} \times CE_i = WSOC_{g,i}\frac{\mu gC}{m^3} \tag{1}$$

$$CE = 1.0273 - 0.8528 \times \exp\left(\frac{-(K_{H,i} - 25)}{244.1}\right) \tag{2}$$

where CE is a unitless collection efficiency expressed as a fraction and c_i is the CMAQ-predicted mixing ratio in ppb_V, multiplied by the ratio of moles of carbon per mole of organic gas i. The unit of $WSOC_g$ is $\mu g\ C\ m^{-3}$. Further details on dimensional analysis concerning units for pressure, temperature, and ideal gas constant are available in the SI. We compare statistical correlations among measured $WSOC_g$ with measurements of chemical species and temperature to correlations among model species for August 2016.

The measured and modeled NO_2 and O_3 are paired in space and time and also compared as mass concentrations by using WRF-predicted temperature and pressure to be consistent among all evaluated gases. Statistical tests, including the Wilcoxon rank sum test to account for non-normally distributed data, are performed between the modeled and measured $WSOC_g$. Pearson correlation coefficients (r) are calculated to determine the statistical associations between measured and modeled $WSOC_g$ and other variables of interest, e.g., NO_x, O_3, and temperature. Statistical tests are performed on the entire data set rather than the averages for each time category. Confidence intervals of 95% are determined via kernel smoothing and bootstrapping.

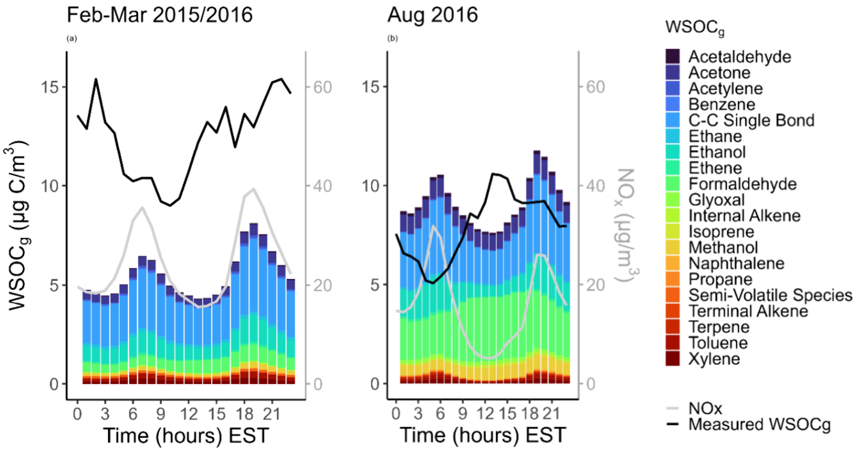

Figure 2. Average monthly diurnal profile of MC collected WSOC$_g$ in (a) February−March and (b) August. The stacked bars represent the individual model species, the black trace indicates the measured WSOC$_g$, and the light gray trace represents modeled NO$_x$ scaled to the right axis. Time is local standard time.

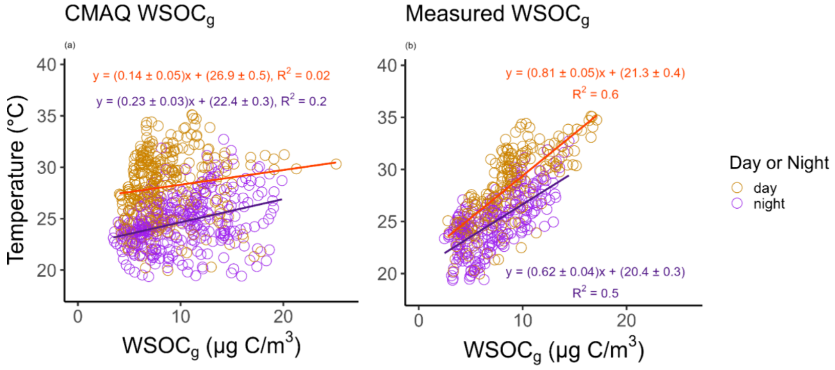

Figure 3. (a) CMAQ-simulated MC WSOC$_g$ vs temperature and (b) measured WSOC$_g$ vs CMAQ temperature in August 2016.

■ RESULTS AND DISCUSSION

The measured and modeled WSOC$_g$ mass concentrations exhibit opposite seasonal patterns, and the diurnal profiles are poorly described by CMAQ. The measured WSOC$_g$ is generally 60% higher during Feb−Mar compared to August values (Figure 1 and Figure S1, Table S3). CMAQ's predicted WSOC$_g$ mass concentrations from simulated MC sampling are generally a factor of 2 greater in August than for Feb−Mar. Similarly, CMAQ's MGLY$_{equiv}$ is more abundant during August (Figure S2). The measured diurnal profile of WSOC$_g$ mass concentrations exhibits the maxima in the afternoon and at night, consistent with the observed diurnal patterns during previous summertime MC sampling in Atlanta, GA and Pasadena, CA.[35,56,71] The simulated MC sampling of CMAQ's atmosphere does not represent diurnal patterns well in any month. When CMAQ's organic gas inventory is expressed as MGLY$_{equiv}$, the diurnal pattern is qualitatively consistent with the WSOC$_g$ measurements for August. The temporal pattern in MGLY$_{equiv}$ is driven by two predominantly secondary species, glyoxal and formaldehyde (Figure S2). Each species has a CE of 1 in the MC sampling. Normalization for MGLY$_{equiv}$ amplifies glyoxal abundance by a factor of 11 and reduces formaldehyde by 10%.

WSOC$_g$ during February−March. During February−March, WSOC$_g$ measurements are up to a factor of 3 greater than the simulated MC samples derived from CMAQ predictions, and paired measurement−model values are significantly different at each hour of the day (Figure 1; Tables S2 and S3). No individual model species in the simulated MC collection exhibits a diurnal profile similar to WSOC$_g$ measurements during Feb−Mar. CMAQ model species that contribute appreciably to WSOC$_g$ in the MC simulations are a combination of primary VOCs and oxidation products. The five most abundant CMAQ species that contribute to WSOC$_g$ mass concentrations during these months are carbon−carbon single bonds (PAR) (45%), ethanol (ETOH) (16%), formaldehyde (FORM) (11%), acetone (ACET) (8%), and toluene (TOL) (4%) (Figure 2).

WSOC$_g$ during August. Every species that contributes to WSOC$_g$ mass in the simulated MC sampling of CMAQ exhibits greater mass concentrations during August (Figure 2), in contrast to the seasonal pattern in measurements at UMBC. During August, the most abundant CMAQ species that contribute to WSOC$_g$ are carbon−carbon single bonds (PAR) (30%), formaldehyde (29%), ethanol (13%), acetone (9%), and methanol (MEOH) (6%). Model bias and absolute differences between measurements and simulated WSOC$_g$ are less during August, yet the diurnal profiles still show key differences in the model representation of the temporal evolution of oxygenated organic compounds. At UMBC, the maximum concentration of measured WSOC$_g$ occurs at 13 EST and corresponds to a local minimum in the CMAQ predictions. The measured minimum at 5 EST corresponds to a local maximum in the predicted values. Formaldehyde is the only model species that exhibits a diurnal pattern similar to the measured WSOC$_g$. Formaldehyde has primary emission sources, but it is primarily a secondary pollutant produced through photochemical processes.[72] The similar diurnal profile of $WSOC_g$ and predicted formaldehyde may suggest that the measured $WSOC_g$ mass is predominantly secondary. Co-located and detailed chemical characterization of $WSOC_g$ measurements is a needed future research direction.

$WSOC_g$ Associations and Overall Model Predictive Skill. During August, the measured $WSOC_g$ and temperature exhibit a significant positive correlation ($r = 0.7$, $p \approx 0$) with different quantitative relationships for day and night (Figure 3). This may reflect temperature-dependent emission differences in VOCs that oxidize to form $WSOC_g$. For example, summertime emissions of highly reactive biogenic VOCs that oxidize to form water-soluble compounds exhibit temperature dependence, increasing with higher temperatures.[53,73] VOC emissions from gasoline evaporation and cold starts of internal combustion engines also exhibit temperature-dependent emission rates that can vary by orders of magnitude.[53,74] Different time-of-day associations between $WSOC_g$ and temperature are also consistent with evolving chemical regimes over a day. The predominance of different oxidants during daylight (e.g., $^\bullet OH$) versus nighttime (e.g., nitrate radical, $^\bullet NO_3$) hours is discussed as a plausible contributing explanation or the measured temporal pattern of $WSOC_g$ in Atlanta, GA.[35] CMAQ $WSOC_g$ predictions do not exhibit a significant association with the temperature during local daylight or nighttime hours. Organic emissions from both biogenic and controllable sources are potential contributors to this model error.[75]

CMAQ-predicted $WSOC_g$ mass concentrations are positively associated with modeled NO_x and SO_2 during all sampling months (Figure 2; Figure S3). Each species exhibits two daily peaks, consistent with the NO_x patterns observed in urban areas.[76−78] SO_2 predictions do not evaluate well against measurements (Figure S4c), possibly due to emissions sources[79] that are unlikely strongly related to VOC sources and less useful to interpretation of $WSOC_g$ predictions. NO_2 predictions evaluate reasonably well at the nearby PAMS site ($R^2 = 0.5$, $p \approx 0$) (Figure S4b), and the relationship between NO_2 and total VOCs is consistent in both CMAQ and measurements (Figure S7). The CMAQ-predicted $WSOC_g$ and NO_x near UMBC exhibit peaks in each season near 5 EST and 20 EST and exhibit significant positive correlations in Feb−Mar ($r = 0.9$, $p \approx 0$) and August ($r = 0.6$, $p \approx 0$) (Figure S5, Table S4). $WSOC_g$ and NO_x "hotspots" are colocated throughout the model domain (Figure 4). CMAQ-predicted and measured primary VOCs also exhibit positive associations with rush hour peaks that are consistent with other IOPs.[80,81] Yet, the measured $WSOC_g$ does not exhibit "rush hour" peaks[35,71] and is out of phase with NO_x, suggesting that the correlation between $WSOC_g$ and NO_x in the model is not representative of actual ambient associations. Hogrefe et al. found that uncertainty in biogenic emissions is associated with significant differences in CMAQ model response to perturbations in anthropogenic NO_x emissions.[39] This is consistent with the predicted model sensitivity of $WSOC_g$ with NO_x not found in observations in this work. Quantitative and detailed understanding is difficult given the lack of speciated measurements

The CMAQ predictions of gas-phase VOCs that form $WSOC_g$ on gas-phase oxidation do not evaluate consistently relative to measurements at the nearby PAMS. Ethane and propane are substantially underpredicted, while acetylene and isoprene are underpredicted (Figure S6a,c−e). Evaluation of benzene and toluene is relatively better; however, all gas-phase organic species have low R^2 values (<0.3) (Figure S6b,f). Total VOCs evaluate similarly to benzene and toluene, while total $WSOC_g$ is underpredicted like its precursors (Figure S8). CMAQ predictive skill for O_3 is relatively excellent in comparison ($R^2 = 0.6$) (Figure S4a,b).

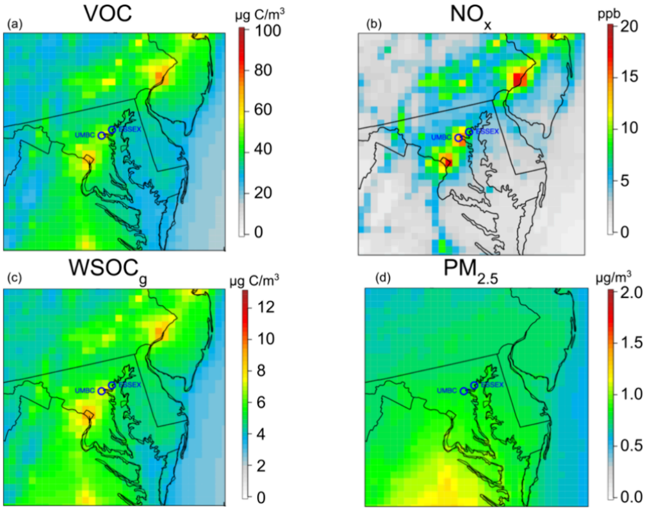

Figure 4. 2016 annual average CMAQ-predicted (a) VOCs, (b) NO_x, (c) $WSOC_g$, and (d) $PM_{2.5}$ for the modeling domain with the Essex and UMBC locations highlighted.

■ CONCLUSIONS

In this novel evaluation of CMAQ-predicted $WSOC_g$, our analysis suggests poor predictive skill for $WSOC_g$ in Baltimore during February−March and August. The absolute values and diurnal and seasonal patterns are not well described. Meteorology is a possible source of error,[46] but CMAQ's predictive skill for O_3 and NO_2 is strong relative to the evaluation of $WSOC_g$. Inadequate description of controllable gas-phase organic emissions, an identified shortcoming of current national emissions inventories[75] in addition to uncertainty in biogenic emission estimates,[39] contributes to measurement−model discrepancies. Description of the chemical oxidation and inadequate surrogate mapping of solubility[82] are plausible contributing factors to poor model performance for $WSOC_g$. The diurnal pattern in $MGLY_{equiv}$, which is dominated by secondary species, exhibits a more similar average diurnal profile to the $WSOC_g$ measurements in August. Neither $MGLY_{equiv}$ nor simulated $WSOC_g$ adequately describes the observations in Feb/Mar. Attainment of the ozone National Ambient Air Quality Standard (NAAQS) plays a determining role in air quality management,[83] as well as development of modeling tools such as CMAQ.[84] Poor model skill for gas-phase organic species, coupled with reasonable representation for O_3 and NO_2 is consistent with an O_3 optimization bias. Such bias can contribute to uncertainty in SOA mass predictions, especially for aqSOA where water-soluble organic gases are the key precursors. Future work to chemically characterize $WSOC_g$ is warranted.

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■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsearthspacechem.4c00379.

Equations used in MC simulations, CMAQ species, statistical analysis, and $MGLY_{equiv}$ (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Annmarie G. Carlton − *Department of Chemistry, University of California, Irvine, California 92617, United States;* orcid.org/0000-0002-8574-1507; Email: agcarlto@uci.edu

Authors

Ellie N. Smith − *Department of Chemistry, University of California, Irvine, California 92617, United States;* orcid.org/0009-0002-1301-8616

Kirk R. Baker − *Office of Research and Development, U.S. EPA, Research Triangle Park, North Carolina 27711, United States;* orcid.org/0000-0003-4417-6799

Marwa M.H. El-Sayed − *Department of Civil Engineering, Embry-Riddle Aeronautical University, Daytona Beach, Florida 32114, United States;* orcid.org/0000-0002-8340-2624

Christopher J. Hennigan − *Department of Chemical, Biochemical and Environmental Engineering, University of Maryland, Baltimore County, Baltimore, Maryland 21250, United States;* orcid.org/0000-0002-2454-2838

Simon Rosanka − *Department of Chemistry, University of California, Irvine, California 92617, United States;* Present Address: Institute of Climate and Energy Systems, Troposphere (ICE-3), Forschungszentrum Jülich GmbH, Jülich, Germany; orcid.org/0000-0001-5929-163X

Complete contact information is available at: https://pubs.acs.org/10.1021/acsearthspacechem.4c00379

Author Contributions

M.M.H.E.-S. conducted mist chamber sampling under the supervision of and with assistance from C.J.H. K.R.B. conducted CMAQ simulations. S.R. assisted E.N.S. with interpretation of CMAQ simulations. E.N.S. performed all calculations presented here and wrote the manuscript under the supervision of A.G.C., who conceived the project. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Notes

The authors declare no competing financial interest.

The views expressed in this paper are those of the authors and do not necessarily represent the views or policies of the U.S. EPA. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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