

Interactive comment on “Overview and evaluation of the Community Multiscale Air Quality (CMAQ) model version 5.1” by K. Wyat Appel et al.

Anonymous Referee #1

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This paper describes recent updates to CMAQ, a chemical transport model used for regulatory and research purposes. The topic of the paper is certainly suitable to GMD and will likely be useful to users of CMAQ. However, the manuscript needs to be improved to better communicate the changes in the code and remove apparent contradictions. In particular, I found the discussion for some of the updates to be too general and did not include sufficient citations justifying those updates.

Major Comments:

1) In general, I found the discussion for some of the updates to be too general and did not include sufficient citations justifying those updates as indicated by some of my specific comments below. In addition, the end of Section 1 and Section 2 need to be reordered to have common steps to improve the clarity of the text. I also have concerns

regarding how the two versions of CMAQ are compared given other differences in WRF and the emissions. I understand that there are often complicating factors that make a more fair comparison possible. Some discussion is included to state why those differences in the configuration arise, but those points could have been made more clearly.

Response: Hopefully we've addressed most of this concern by addressing the specific comments below. In general however, we tried to include enough detail so that the reader understood what basic changes were made and why. If the reader wishes to get more detailed specifics of the changes made, they are referred to the technical documentation for the model release.

2) Section 5.2: I like evaluating the models using profiles from the DISCOVER-AQ data, since observations at the surface only provide a small slice of the atmosphere. However, Section 5.2 seems rather brief and overly simplistic. Extensive measurements were collected during the campaign I presume, yet only one profile is shown. It does illustrate the differences between models, but only for one case. The authors needs to either delete the section, provide a more extensive evaluation, or justify why only one profile is needed. One way to summarize the aircraft data is to showing percentiles of both model and observations as a function of height. In addition, why not use the NASA lidar data to illustrate differences in PM? The authors describe changes in aerosol optical properties but do not evaluate this part even though data is available to evaluate the optical properties.

Response: Since the objective of this section is to evaluate the change in model performance for NOY, AN and PNs, the section has been retitled to reflect its purpose and not suggest to the reader that this section will be a comprehensive evaluation against aircraft measurements. Several statistical metrics of NOY performance have also now been included in the section to help expand the analysis provided and highlight the greatly improved performance of NOY in CMAQv5.1. While it would be nice to be able to show additional profiles from other days and include measurements from other networks, the point of the section was to simply inform the reader of the large improvement in NOY performance and an example of the change in ANs and PNs mixing ratios that can be expected in the new model. Future evaluations of CMAQ will focus specifically on the DISCOVER-AQ time period and utilize the measurements made to a much greater extent.

Specific Comments:

Page 1, line 27: Does "thereby reducing the PM_{2.5} bias" refer to the previous bias? Please be specific.

Response: Added a statement indicating underestimation of PM2.5 by CMAQ in the summer to clarify what bias is being reduced.

Page 1, lines 28-29: The text in these two lines seem to contradict one another in terms of the PM2.5 bias.

Response: Clarified that this refers to the consideration of the effect from all the changes made to the model and not just a single update as previously referred to.

Page 1, lines 31-32: Line 31 says v5.1 has a higher O3 bias, but in next line says error is better. I can understand that the correlation could be better in v5.1 even though the bias is worse, but the authors are not clear what they mean here.

Response: Corrected text to read that only the correlation improved and not the error.

Page 1, line 36: What does “significantly” really mean? This is not specific. Page 2, lines 20-24: Consider making this text a bulleted list.

Response: Removed the word significantly since it is subjective. Opted to avoid introducing bulleted text into the manuscript.

Page 2, lines 18-29: I was trying to relate the changes described in this paragraph, with Section 2. But upon first reading the paper, it was not clear to me that these two parts were necessarily referring to the same changes. The text could be improved if lines 18-29 were written to be parallel to Section 2, or visa versa.

Response: Reordered this paragraph to make it consistent with the order in which the model updates were presented in section 2.

Page 3, lines 18-24: Please include a reference justifying the revised stomatal conductance. As written, it seems the modification is simply a tuning parameter that improves some quantities in the predictions. There could be easily other changes in the model that could improve the quantities that were mentioned.

Response: A reference was added regarding the origin of the stomatal conductance values.

Page 3, line 25: Mention values of the heat capacity used in the old and new versions.

Response: These values were added to the text.

Page 3, line 32: So what is the Pr now? The authors say they changed it, but are not specific about this parameter.

Response: The Pr is (and was) a function of the eddy diffusivity values of momentum and heat. Previous, these values were the same and there Pr was always equal to unity. That is no longer the case.

Page 3, line 32-36: Similarly, what are the new stability functions and are there some published results to describe them?

Response: The stability functions are described in Pleim et al. 2016 which is provided as a reference in this section.

Page 4 line 7, This line is confusing. I am missing some details since the phrase “little difference between the initial MOL estimate and the final re-calculated value” is basically saying the code will do nothing. Then what is the point of the code? To me the test associated with MOL assumes the reader is already somewhat familiar with this subject, but I do not understand the logic here.

Response: This statement was removed from the text as it was unnecessary and requires a greater understanding of the use of the MOL value in CMAQ and WRF that described in the text.

Page 4, lines 9-16: The text on gravitational settling seems out of place in this section.

Response: This description has been moved to the end of Section 2.5.

Page 4, line 26: There are many studies, not just the two cited, that indicate IVOCs are missing in the emission inventory. Suggest changing text to include “e.g.” or “for example” or something similar.

Response: Changed the text to indicated the provided references are examples.

Section 2.2: In general, I found the text in this section to be confusing in terms of what is actually new in v5.1 compared to older versions. The level of detail is rather minimal.

Response: The goal of this section is to provide an overall understanding of what was updated and why. It is not intended to document in detail every change made to the aerosol code in CMAQ. Text was added to the beginning of the section that points the reader to the CMAQv5.1 technical documentation which includes in detail all the changes made to the model.

Page 5, line 15: What is “more consistent” mean? Why isn’t it completely consistent?

Response: We have revised the paragraph to better define what is and is not consistent with the meteorological model. The updates in photolysis calculations in CMAQ v5.1 related to clouds were intended to ensure internal consistency between cloud mixing, aqueous chemistry and photolysis. The reason cloud treatment in CMAQ is not currently “completely consistent” with WRF is the way that sub-

grid convective clouds are handled. The sub-grid convective cloud scheme in CMAQ, which is responsible for convective transport of chemical species, aqueous chemistry, and wet scavenging, is a simple bulk scheme based on the convective cloud model in the Regional Acid Deposition Model (RADM; Chang et al., 1987) but with convective transport based on the Asymmetric Convective Model (Pleim and Chang, 1992). Since the CMAQ cloud scheme uses the convective precipitation rate to diagnose sub-grid mass fluxes, the location and timing of precipitating convective clouds are consistent with WRF. A new convective cloud scheme for CMAQ based on the Kain-Fritsch scheme in WRF is currently being tested to improve consistency across chemical and meteorological components of the system.

Page 5, lines 20-21: What does “run time options” mean? I assume the authors mean the user has the ability to choose these options. “run time options” sounds like unnecessary jargon.

Response: We removed the term and added text on how a user may use either of the two options.

Page 5, lines 24-25: This sentence does not describe how “cloud fraction, sub-grid cloud fraction, resolved cloud water content” are actually used.

Response: These parameters are simply provided in the text as examples of new diagnostic values that are available as output in the new version of the model in case a user wishes to examine them. They are not new variables used in the CMAQ model. We also added text to the paragraph briefly describing the calculation method for photolysis rates and how the clouds contribute to the calculation.

Page 5, line 27: The satellite data can be used to evaluate clouds, but it cannot be used to directly evaluate photolysis calculations. The authors need to be more specific here. I think the authors mean that the clouds indirectly determine where photolysis rates may be high or low, but the satellite does not provide any quantitative estimate of photolysis.

Response: We agree and revised the paragraph to state this point. Note that this paragraph was also moved to section 4.3 since the focus is on model evaluation. The revised sentences now state:

“Additional diagnostic evaluation of photolysis/cloud model treatment in CMAQ was conducted based on the model predicted cloud albedo at the top of the atmosphere. The predicted cloud albedo from WRF3.7, CMAQv5.0.2 and CMAQv5.1 were evaluated against cloud albedo from NASA’s Geostationary Operational Environmental Satellite Imager product (GOES; <http://satdas.nsstc.nasa.gov/>). This evaluation was used to qualitatively determine if one CMAQ version better considers how clouds affect calculated photolysis rates.”

Page 5, line 34: Do you mean photolysis rates at the surface? Please be specific. Surface values will differ from those aloft.

Response: The figure shows what the cloud parameterization between version 5.0.2 and 5.1 implies about the cloud albedo or reflectivity at the top of the atmosphere. Changes in photolysis rate are integrated over the vertical column so the paragraph does not discuss photolysis rates at a specific altitude. Also, the revised text attempts to better explain the analysis and displayed results.

Page 5, lines 37-38: This statement is about the clouds, but c) and d) are about photolysis rates. I understand the photolysis rates reflect the cloud distributions, it is just strange the way the sentence is stated. As I said before, the use of “more consistent” leads me to wonder in what ways the clouds in WRF and CMAQ still differ. What are those ways?

Response: We revised the paragraph in section 2.3 to better explain how WRF and CMAQ differ in the cloud description, specifically the sub-grid or convective clouds (please see response to previous question for more details). The paragraph describing the figure has been moved to section 4.3. The figure is now referred to as Figure X and the statement in question has been reworded to more accurately describe that what is being plotted is based on the cloud parameterization in the CMAQ system, not just within the photolysis module:

“Figure X shows the average cloud albedo or reflectivity at the top of the atmosphere during daytime hours in July 2011 derived from the GOES satellite product (5a), and the cloud parameterizations within: (5b) WRF3.7, (5c) CMAQv5.1_RetroPhot and (5d) CMAQv5.1_Base.”

(Note that the “CMAQv5.1_RetroPhot” and “CMAQv5.1_Base” abbreviations have been defined at the beginning of section 4.3.)

Page 6, line 6: How was “most important” determined? More important than what? It seems that the modifications are being added based on recent research activities, but it is not clear why these are more important than other new pathways that may have been reported in the literature. Please explain.

Response: Since the term “most important” is inherently subjective, we opted to remove that statement since it was not important to the discussion regarding the mechanism updates.

Page 6, line 12: N and Fc need to be defined.

Response: The statement referencing these values has been removed since it's not critical at all to discuss these variables and they are described in the citation provided.

Page 7, lines 6-9: This text is really only saying that some updates have been made, but gives no real specifics on what those updates actually are. How will this help users?

Response: The specifics of these updates are providing in the technical documentation available through the CMAS website. A link to the documentation is provided in the beginning of the section. Here we're making the reader aware of the changes and why they were made.

Page 7, line 26: The text mentions overestimates of biogenic VOCs at coastal sites, but this sentence seems to require a reference to know what study pointed that out and how.

Response: The text has been modified to better explain how the overestimation was determined and addressed, and now also includes a reference.

Page 8, Section 3: What is missing from this section is a list of parameterizations used in WRF.

Response: Modified the text to include the specific parameterizations that were employed in the WRF simulations. In addition, now include the WRF namelists in the supplemental material.

Page 8, line 14: I am not sure why the same version of WRF was not used to drive the two versions of CMAQ. I assume it is to have their older treatments in the land-surface and PBL parameterizations; however, there are likely other changes in the model as well that could cause differences. Please comment, and I think it is worthwhile to reiterate at this point why the two versions of WRF are used.

Response: The text was modified to explain why different versions of WRF were used. In short, because the updates made in WRFv3.7 were tied to similar updates made in CMAQv5.1, those two version of the models need to be used together (without modifications to the MCIP code). Similarly, WRFv3.4 is tied to CMAQv5.0.2 without additional modifications to the MCIP preprocessor. Hopefully this is now clear to the reader in the main text.

Page 8, lines 19-39: I am also confused why different emission inventories are used. This will drive differences in the v5.0 and v5.1 simulations that are beyond just the changes to the parameterizations.

Response: A new emissions platform became available after the v5.0.2 simulations were complete. It was felt that in order to obtain the best model results the latest emissions platform should be used and therefore was used for the v5.1 simulations. Sensitivity tests were performed to assess the impact that the changes in the emissions platform had on the model results and the impacts were determined to be small. A figure showing the impact of the emissions platform change on ozone and PM2.5 in January and July has been added to the text to quantify to the reader the impact from the emissions platform

change.

Page 10, lines 27-28: While I cannot disagree with these sentence, I think the explanation is rather simplistic. SOA depends on photochemistry and has been shown to be correlated with O₃. So if O₃ increases, one could expect increases in SOA and therefore an increase in PM_{2.5}.

Response: Added a statement regarding how the change in oxidant concentration could impact the formation of SOA and therefore PM_{2.5} concentrations.

Page 10, line 39: The authors note that the total concentration of the new SOA species are small. One could conclude here that why were they included in the first place? It would be useful to reiterate that the PAH species are for health reasons and will need to be evaluated in the future. I am less sure about the ALK species. In section 2, the authors not that only the “most important” changes are made, but it is not clear why this is important.

Response: Statements were added to indicate that while the overall monthly difference in concentration of these species is small, the episodic isolated concentration can be higher. Also indicated the importance of these species in health related studies.

Page 11, lines 1-10: Was the temperature the same between the two versions? Since biogenic emissions are temperature dependent, I am wondering how much difference here is due to meteorological effects versus the changes in chemistry.

Response: The temperature difference between the two simulations is very small due to the use of four-dimensional data assimilation in the WRF simulations and does not affect the biogenic emissions significantly.

Page 11, lines 30-31: Here is a first mention that CMAQ produces more clouds than WRF. The reason for the differences would be useful to describe in Section 2. I still do not understand why CMAQ would have a different representation of clouds, which can only complicate interpretation of the effects of clouds on chemistry. Later in lines 37- 38, they mention differences in sub-cloud treatments. Again this should be stated more upfront in the text. Why is it difficult to have consistent treatment of clouds between the models?

Response: Moved this paragraph to Section 2.3 as it seemed more appropriate there. Greater effort has been made in that section to explain why the clouds in WRF differ from the clouds in CMAQ.

Page 11, line 36: The authors mention “WRF cloud parameterization” but they should specifically state in their model set up which microphysics and cumulus parameterization they used. The way the text is stated, it implies

WRF has only one when in fact there are many options. It is not clear that the underprediction in clouds they have could have been fixed or improved using another choice of microphysics or cumulus parameterization.

Response: The text now includes which WRF parametrizations were used in the simulations. In addition, the WRF namelists used have been added to the supplemental material.

Page 12, line 19: The second phrase of this sentence is redundant with the first phrase and adds no new information; therefore, it should be deleted. For the same reason, the second phrase in the sentence in lines 20-22 should be deleted.

Response: The redundant lines were removed.

Page 12, line 28, the authors mention low (I assume lower) PBL heights. So the difference in O₃ are driven by the differences in meteorology and it would be useful to quantify this difference in PBL height. If the difference in PBL is on the order of 10's of meters, how does that compare to the vertical grid spacing of the model to actually make a difference?

Response: This was to note the relatively low PBL heights that typically occur over water versus land and as a result the change in ozone precursors can amplify the change in ozone due over water to low PBL heights. This is not a statement of the actual difference in PBL height between the two WRF simulations.

Figure 5, Perhaps it would be more useful to use percentage changes instead?

Response: As a compromise, added the approximate percent change in O₃ and PM_{2.5} to the text describing Figure 5.

Section 4.4: The differences described in this section seem small, so how does this demonstrate a major update of the code? Does this mean the code changes are important theoretically, but they do not make big difference in the predictions.

Response: Some of the differences are actually quite large for monthly averages. However, the changes made to the atmospheric chemistry are incremental in nature.

Page 12, line 32: I am thrown a bit by the phrase "operational performance". "operational" may mean different things to different communities. Section 4 had a comparison of the models, which is repeated here but now include observations. Maybe just say the performance is evaluated by comparing the models with one another and observations?

Response: Removed the word "operational" as this was a source of unnecessary confusion without the

appropriate context.

Page 13, line 37: Is the change statistically significant?

Response: Statistical significance doesn't apply in this case since we're comparing two model simulations against the exact same set of observations. So, any change is by default statistically significant. However, it would remain in the hands of the reader to determine whether the change is significant to their application.

Page 14, lines 10-11: The authors note an improvement in certain aerosol species, yet emission are different between the simulations. On the next page on line 16, they mention the differences are due to emissions. Why is this then important in terms of the code changes in CMAQ?

Response: I don't actually see where in line 16 it is mentioned that the differences are due to emissions. It is stated that the difference are primarily the result in differences in the concentration of primary emitted species, but that it likely the result of changes in the meteorology (lower or higher PBL heights) and changes in the emissions.

Page 15, line 22: Abbreviations are used for states here, but not elsewhere so there is an inconsistent use. I suggest writing out all state names since international readers will not necessarily know what the state abbreviations are.

Response: Abbreviations are no longer used for the state names.

Page 18, line 32: References to papers in preparation should not be included. Are there other references that can be used?

Response: This paper is actually referenceable as it is online and includes a doi. The text has been changed to no longer indicate the paper as in-preparation.

Page 18, line 33: This section is titled "discussion" but this section contains little new discussion regarding the model results. It reads more like a summary section.

Response: The title of this section has been changed from "Discussion" to "Summary" as it does constitute a summary of the work and not a new discussion.

Review of the manuscript “Overview and evaluation of the Community Multiscale Air Quality (CMAQ) model version 5.1

This article describes the changes that have been brought to the CMAQ model from version 5.0.2 (released in April 2014 according to the CMAQ website) and version 5.1, released in Dec. 2015, including comparison of model performance between these two model versions, mostly for ozone and PM_{2.5} over the continental United States. Five different simulations have been performed for the year 2011 (or two months in this year) to evaluate the changes in model performance due to the global and simultaneous upgrade of WRF and CMAQ version as well as of the emission database, but also to separate the contribution of different changes in the models. The authors show in a convincing way that the improvement between model v. 5.0.2 and v5.1 is substantial, even though it is unclear whether this improvement could be due in part or totally to the change of emission datasets. The sensitivity of versions 5.0.2 and version 5.1 of this model to emission reduction scenarios is also evaluated in terms of RRF (relative response factor) for ozone, and of emission cut simulation / base simulation (for PM_{2.5}).

This article is definitely within the scope of GMD. The improvements in CMAQ that are presented seem substantial even though for some of them the detailed explanation of what has actually been done and why lacks detail. A considerable work of validation has been performed.

Many aspects of the manuscript need to be improved before final publication can be considered. These aspects include the traceability / reproducibility of results (exact description of model), and providing a real and complete model overview. Also, the possibility that the described improvements in model performance are due totally or partly to the use of a new emission dataset must be ruled out.

Less importantly, a clarification of the links between WRF and CMAQ is needed (all these points are developed below in points GC1 to GC6 of my review which I think should definitely be addressed).

This article has the potential to be read and cited by many researchers or operational modellers that use CMAQ for their studies, operational previsions, and/or contribute to its development. It also reflects a huge amount of work by the CMAQ development team. This is why, **on the one hand, I recommend that this study shall be published in GMD, but I think that some important aspects of the paper definitely need to be improved before publication in GMD, including the detailed description of the model parameterizations that are changed (traceability) and a complete overview of CMAQ v5.1.**

The article is clearly written and the language level is good as far as I can say.

General comments

GC1 Sensitivity to emissions

It is appreciable that sensitivity simulations are performed to evaluate the impact of the various changes between the v5.0.2 setup with WRF 3.4 and NEIv1 emissions and the v5.1 setup with WRF 3.7 and NEIv2 emissions.

However, I have the feeling that, if the idea is to test the sensitivity of the results to the various improvements performed, then a simulation with the v5.1 setup but with NEIv1 emission dataset should be provided. In the present version, all simulations with v5.1 are performed with NEIv2 emissions and all simulations with v5.0.2 are performed with NEIv1 emissions. So the improvement between both model versions, which is shown in a convincing way, could after all be due in part or totally to the improved emission dataset. A sensitivity experiment to emissions is in my opinion needed to rule this hypothesis out and show in a direct way that the improved results are really due to the improvements in WRF and CMAQ and not to better emission input datasets.

I feel that a convincing answer to this caveat needs to be brought before publication. Otherwise, the reader has no proof at all that the described improvements are not just an effect of changing the input emission dataset

Response: A new emissions platform became available after the v5.0.2 simulations were complete. It was felt that in order to obtain the best model results the latest emissions platform should be used and therefore was used for the v5.1 simulations. Sensitivity tests were performed to assess the impact that the changes in the emissions platform had on the model results and the impacts were determined to be small. Obviously it was not made clear in the manuscript that the overall impact from the emission platform change was small. Hopefully this is now made clear in the text. In addition, a figure showing the impact of the emissions platform change on ozone and PM2.5 in January and July has been added to the text to quantify to the reader the impact from the emissions platform change.

GC2: reproducibility/traceability, precise description of parameterizations

In some occasions, statements are done that some parameterizations have been “improved”, or “changed”, but failing to describe exactly what has been changed in which way, questioning the reproducibility of the results (see below comments C7, C10, C14). Details, references and, if necessary, equations need to be brought so that developers of other models are able to test similar changes in their models.

Point 6 in the GMD review criteria states: *“In the case of model description papers, it should in theory be possible for an independent scientist to construct a model that, while not necessarily numerically identical, will produce scientifically equivalent results. Model development papers should be similarly reproducible. For MIP and benchmarking papers, it should be possible for the protocol to be precisely reproduced for an independent model. Descriptions of numerical advances should be precisely reproducible”*.

-> The present manuscript clearly fails to meet this criteria. This is not a reason for rejection because the authors will easily be able to correct this caveat in the review process, but this should definitely be done before final publication.

Response: The details regarding the parameterizations and options employed in both the WRF and CMAQ simulations has been expanded in Section 3. In addition, the namelists for the WRF simulations have been included in the supplemental material. As for the CMAQ model, the model code for all the versions of the model presented here is available for download through the CMAS Center website. The code is open source and freely available. The input data, including the emission and MCIP (WRF) data are all available upon request from the corresponding author. Even the output from any/all the model simulations performed here are available upon request as well.

GC3: Need for a real model overview

The title of the paper is “Overview and evaluation of the Community multiscale Air Quality (CMAQ) model version 5.1” but in my opinion the paper clearly lacks an overview of the model. I think that a Section “Overview of CMAQ 5.1” or similar should be introduced between the Introduction and the

section about the scientific improvements.

This section should include at least the basic information one would expect to find about a chemistry-transport model: since when has this model been developed? What kind of grid does it use? With what type of transport scheme (horizontal and vertical transport), is the transport Eulerian, Lagrangian, mixed? What is the recommended range of use in terms of resolution, domain size, regions of use, vertical extension? What are the inputs that are needed and the variables that are provided as an output? Are the aerosols treated in a sectional or modal way, and which physico-chemical processes are included regarding the aerosols? For example, we read that gravitational sedimentation is now included but we do not know if and how processes such as evaporation, coagulation, dry and wet deposition etc. are treated.

Maybe the authors consider that the focus of this article shall be uniquely the increment from version 5.0.2 to version 5.1 instead of a real model overview, but in this case the title would have to be changed accordingly (and the interest of the paper would be greatly lessened in my opinion, questioning the interest of the publication). In the present state, the paper does not reflect the title, because it does not include an overview of v5.1, only a set of sensitivity studies between v5.0.2 and v5.1.

Response: It's true that this manuscript is intended to provide a description of the changes that were made between CMAQ versions 5.0.2 and 5.1, along with a comparative evaluation of the results from the previous version of the model and the latest version. It is not intended to be a true "overview" of the modeling system. There have been several papers published that present a general overview of the CMAQ model from a physics and functionality perspective (Byun and Schere, 2006 for example, which is referenced in Section 1). The usefulness of presenting results from one model version to another can in some cases be questionable, however in this case it was felt that the scope of the updates made to the modeling system warranted informing the user community of the model updates and providing evaluation results that users of the model can use to determine whether or not that want to use the new model for their applications. As for the title of the manuscript, it was changed to better reflect that this article is a description of the changes in and evaluation of the CMAQv5.1 model, and not an overview of the CMAQ modeling system.

GC4 Need for a general presentation of model outputs

I feel that the reader lacks a spatialized vision of the model outputs and their characteristics compared to the known features of atmospheric composition over north America. It would be very helpful to provide a map of simulated ozone for the months of January and July, as well as simulated NO_x, PM_{2.5} for these months with v5.1, possibly superposed with the measured average where station data is available, or any other way to give a spatialized vision of model outputs in comparison with state-of-the-art knowledge of the atmospheric composition over the continental US.

Response: This is a good suggestion. At the risk of inundating the manuscript with too many figures, we opted to provide seasonal plots of PM_{2.5} and O₃ from CMAQv5.1 in the supplemental material. This should suffice to give the reader the reference that the reviewer is suggesting.

GC5 Better description of modelling setup

The modelling setup (Section 3) should be described more carefully. Very little space is dedicated to describing the CMAQ configuration for the main simulation, and this section mostly describes the changes between NEI emissions v1 and v2. I think that some information is clearly missing: what kind of initial and boundary conditions are used for the main species, what advection schemes, and the main user options that have been chosen for the simulations. This is particularly the case as CMAQ is a very modular model in which many choices are left to the user, as stated on other CMAQ-related documents.

The same applies to the WRF configuration, particularly as WRF seems very intricate with CMAQ (it

seems that some of the updates need to be performed simultaneously in WRF and CMAQ). WRF also has many configuration options, and some of them are very critical for chemistry-transport modelling, such as for example the PBL scheme, but also the convection parameterization (if used), the eventual damping options to avoid instability over steep terrain (which is important since the continental US include major mountain ranges), etc. Some information is given later in the text but should be given in Section 3 as well. It should also be mentioned what initial and boundary conditions have been used for WRF, and if some nudging has been applied.

It should also be mentioned explicitly if a spinup period has been performed (and discarded) for each simulation. This is particularly the case for the July and January simulations, which last only one month.

Response: The model setup for both CMAQ and WRF has been expanded to include many of the options employed in both models. The WRF namelists are now provided in the supplemental material as well. Information regarding the spin-up periods used has also been added to Section 3.

GC6: Precisions about the WRF model and its links with CMAQ

In many parts of the CMAQ and WRF seem very intricate (as soon as the abstract, which states “Version 5.1 of the CMAQ model was released to the public which incorporates a large number of science updates (...) These updates include improvements in the meteorological calculations **in both CMAQ and WRF**”. Also on p. 13, l. 8-12 (as well as in the conclusion), we find a sentence that tends to indicate that WRF-CMAQ would even have to be considered as a single “WRF-CMAQ modeling system”, that “therefore should be evaluated together”. This raises some questions:

- * Can CMAQ be used with other meteorological models than WRF (such as reanalysis or outputs from national meteorological centers)? Is it recommended by the CMAQ developers?
- * If these models are so intricate together, would it not be relevant to change the title to include this concept of “WRF-CMAQ modeling system”?

Response: This is good point to be made. While CMAQ can be used with other meteorological models (MM5 for example), it does have strong ties to the WRF model through consistent mixing schemes and land-surface treatments. So, while it’s not truly correct to call it exclusively the WRF-CMAQ modeling system, what was tested and evaluated here was the WRF-CMAQ modeling system.

Minor general comments

- Some parts of the article are not friendly for a non-US reader. For example, the 2-letter codes for US states are not well-known to the international public. Either the authors should provide a map of these codes, also including the five zones defined p. 14, l. 6-8, or give the complete names of the states.

Response: All state names are now spelled out.

- Many long URLs are given between parenthesis in the text (e.g. p. 6, l. 27-28). They should probably be given as footnotes.

Specific comments:

Section2

C1 p. 5, l. 31: Is this time interval valid for all the domain? Days in July should last much longer than

12 hours at least in the north of the domain, and the daytime interval must be very different from the west to the east of the simulation domain (about 5000 km, which is about 4 hours time lag in the solar time). Using points from 11:45 to 23:45 UTC from west to east would result to using data points from mid-morning to the sunset at the eastern part of the domain, and from dawn to mid-afternoon in the west of the domain, which is critical as cloudiness often has a strong diurnal cycle.

I recommend that all the available daytime data points shall be used for this comparison.

Response: All available data from the satellite product are used in the average in Figure 1a (Note this Figure has been moved and is now referred to as FigureXa). The figure in the Supplemental Information section S1 shows the number of daytime hours (11:45UTC – 23:45UTC) with available GOES cloud albedo data during July 1 – July 31, 2011 for the modeling domain. Regions in the eastern half of the US have a larger number of available satellite observations (on the order of 390hrs) compared to the western coast which has < 340hrs. Since the reference to the time window of 11:45UTC-23:45UTC caused unnecessary confusion we have removed this from the main text. We now point readers to the Supplemental Information for further description of the hours of available satellite data:

“The satellite data are available at 15 minutes prior to the top of the hour during daytime hours and were matched to model output at the top of the hour (see section S.1 in the supplemental material for further information).”

C2 p. 5, l. 32: the description of Fig. 1 does not fit that in the caption of Fig. 1 (the latter one seems to be more relevant). The average cloud albedo seems not to be shown.

This should be clarified.

Response: The reviewer is correct. The wrong Figure 1 was included with the original submission of the manuscript. In the revised version of the manuscript this Figure (now called Figure 4) does show the average cloud albedo, consistent with the description in the text. The Figure caption has also been changed to say:

“Figure 4. The average cloud albedo during daytime hours in July 2011 derived from (a) the GOES satellite product (b) WRF3.7 (c) CMAQv5.1 with photolysis/cloud model treatment from v5.0.2 and WRF3.7 inputs (CMAQv5.1_RetroPhot) (d) CMAQv5.1 using WRF3.7 inputs (CMAQv5.1_Base).”

C3 p. 5, l. 36-371: The authors should explain why it is needed to have a convective cloud model within CMAQ and not use cloud fractions and water content provided by WRF, particularly if CMAQ and WRF almost form a single modeling system as stated elsewhere.

Response: We revised the preceding paragraph to explain why CMAQ cannot not use convective cloud predicted by WRF. The updates in photolysis calculations CMAQ v5.2 related to clouds were intended to ensure internal consistency between cloud mixing, aqueous chemistry and photolysis. The reason cloud treatment in CMAQ is not currently “completely consistent” with WRF is the way that sub-grid convective clouds are handled. The sub-grid convective cloud scheme in CMAQ, which is responsible for convective transport of chemical species, aqueous chemistry, and wet scavenging, is a simple bulk scheme based on the convective cloud model in the Regional Acid Deposition Model (RADM; Chang et al., 1987) but with convective transport based on the Asymmetric Convective Model (Pleim and Chang, 1992). Since the CMAQ cloud scheme uses the convective precipitation rate to diagnose sub-grid mass fluxes, the location and timing of precipitating convective clouds are consistent with WRF. A new convective cloud scheme for CMAQ based on the Kain-Fritsch scheme in WRF is currently being tested to improve consistency across the chemical and meteorological components of the system. This future model update will allow sub-grid cloud fraction and water content information from WRF to be used within the sub-

grid cloud-related processes within the CMAQ system.

C4 Fig. 1: The methodology to produce these maps should be precised. Is a threshold placed on cloud albedo to decide that a particular hour is or is not cloudy? Is this threshold the same in the models and for the GOES data?

Response: The wrong Figure 1 was included with the original submission of the manuscript. In the revised version of the manuscript this Figure (now referred to as Figure 4) shows the average cloud albedo (consistent with the description in the text). There is no threshold used or needed with the new figure.

C5 p. 5, l. 37-38: the statement that the model cloudiness is “more consistent with the WRF parameterization” is possibly correct but not shown by the figure, since WRF3.4 cloudiness is not provided for comparison with CMAQ v5.0.2 cloudiness. Also, it is hardly a surprise, since CMAQ v5.1 clouds are produced from WRF 3.7 cloud data it would be alarming if the results are very different.

Actually, this paragraph seems to me a bit tilted towards suggesting that CMAQv5.1 cloudiness is better than CMAQ v5.0.2, but the figure does not allow to make such a statement, since CMAQ 5.1 cloudiness is compared to the cloudiness of WRF 3.7, which is almost the same data, while CMAQ v5.0.2 is compared to the actual satellite data, which is more of a challenge. Actually, in my eyes, visual comparison between Figs 3c and 3d to Fig. 3a shows that, above most of the continental US and the surrounding oceans (except maybe the center-north of the US), CMAQ 5.0.2 cloudiness is in much better agreement than CMAQ 5.1 with the observed cloudiness.

Response: We revised the paragraph in section 2.3 to better explain how WRF and CMAQ differ in the cloud description, specifically the sub-grid or convective clouds. The motivation for the updates in the cloud treatment in CMAQv5.1 is the improved consistency with the WRF parameterizations and this Figure is intended to emphasize this improvement. (Note that the figure in question has been moved to section 4.3 and is now referred to as Figure 4). The cloud albedo in Figure 4c that represents the cloud treatment from CMAQv5.0.2 was based on inputs from WRF3.7, not WRF3.4. This is now made more explicit in the text and the Figure caption to avoid any confusion.

The authors invite to compare Fig. 3c to 3a and 3d to 3b, but I recommend that they also invite the reader to compare Fig. 3d to 3a and explicitly comment the comparison between CMAQ v 5.1 cloudiness to the Goes data and comment why the agreement does not seem as good as with CMAQ v5.0.2 over many areas.

Response: The readers are provided information on the differences between the CMAQv5.1 cloudiness and the GOES data in the following paragraph in section 4.3:

“Two notable issues remain with the v5.1 modeled cloud parametrization. The photolysis cloud parameterization in v5.1 produces more clouds over water compared to the WRF parameterization, which is itself biased high for some parts of the Atlantic Ocean compared to GOES. This issue will be addressed by science updates planned for the CMAQ system and evaluation results are expected to improve in the next CMAQ release (See Section 7.4). A more significant issue, from an air quality perspective, is the under-prediction of clouds over much of the Eastern and West Central US in the WRF predicted clouds, which is now directly passed along to CMAQ. This misclassification of modeled clear sky conditions can contribute to an over prediction of O₃ in these regions. Resolving this issue will require changes to the WRF cloud parameterization. Future research will also include changing the sub-grid cloud treatment currently used in the CMAQ system to be consistent with the sub-grid parameterization used in WRF. Section S.1 in the supplemental material provides a table with additional evaluation metrics

of the modeled clouds over oceans versus over land and also describes how cloud albedo was calculated for the three model simulations.”

Also, it would be interesting to have the same 4 figures shown and commented for the month of January (eventually as a supplement).

Response: We did attempt to make this figure for January but found that the satellite retrieval method for cloud albedo gave unreliable diagnostic information for locations with snow cover.

C6 p. 6, l. 11-12: “the rate constant (...) low-pressure limit”: please clarify, and define what are N and Fc in the subsequent parenthesis.

Response: We opted to remove the statement containing the values for N and Fc, as they are the same as those reported by Bridier et al. (1991) which is referenced in the section. Including the updated values seemed unnecessary given the that they are available in both the cited paper and the CMAQv5.1 technical documentation, and would actually be quite laborious to define here.

C7 p. 7, l. 11-19

The modification to the sea-salt emission schemes is described in a rather vague way. I recommend that the following information is added:

- Give the equations that control the sea-salt emission processes in CMAQ in open ocean and in the surf-zone and the reference from which they were inferred. It would also be helpful to the reader to also show the size distribution of sea-salts in the former and in the new version (“the size distribution (...) was expanded to better reflect ...” seems a bit to vague to me, not permitting reproducibility.

Response: Numerous references were included in this section that contain the information requested above. In addition, the CMAQv5.1 technical documentation also contains detailed information regarding this update to the model. It seems excessive to rehash here the specific equations and values implemented in the new model when they are easily obtained through the citations provided. The combination of the citations provided and CMAQ technical documentation should provide information needed for reproducibility. In addition, the CMAQ code itself containing the updates is available as well.

C8 subsection 2.1, p. 3, seems to address issues about vertical mixing and air-surface exchanges rather than explicit transport. I think it would fit better in the 2.5 subsection.

Response: This section has been moved to the end of Section 2.5.

C9 p. 7, l. 23: Niinements to Niinemets?

Response: The reference name has been corrected and added to the list of references.

C10 p. 7, l. 23-24: “A leaf temperature algorithm was implemented that replaced the 2m-temperature...”. I think there is not enough detail to guarantee reproducibility, or the possible use of this algorithm by other modellers. I recommend that the idea of this algorithm and its equations are given, and if possible that the interested reader shall be oriented to a publication describing this algorithm.

Response: A detailed description of the updated algorithm is available in the CMAQv5.1 technical documentation, which is now provided as a link in the text and the reader is now encouraged to reference for additional details.

C11 p. 7, l. 24-25: it is unclear to me how 2m-temperature can be consistent with emission factor

measurements (2m temperature should be consistent with 2m-temperature measurements...). Please reformulate or clarify this sentence.

Response: This statement has been expanded to make it more clear what exactly was changed to make the calculation in the CMAQv5.1 more consistent regarding the 2-meter temperature and emission factor in BEIS.

C11 p. 7, 25: please define BELD

Response: BELD has already defined in the text as “Biogenic Emission Land-use Data”.

Section 3

C12, p. 8, l. 5: it is interesting that the model is run up to 50 hPa, well into the stratosphere. I think it would be interesting to have a glimpse of the model outputs in the stratosphere (or throughout the whole atmospheric column) and their validity, particularly regarding ozone. For upper troposphere/lower stratosphere, comparison with either satellite data or aircraft data such as MOZAIC (<http://www.iagos.fr/web/>) would, I think bring something useful to this study. Also, it would be interesting to know whether additional reactions are needed in CMAQ to take into account the lower stratospheric chemistry, which is different from tropospheric chemistry.

Response: It's really beyond of the scope of what this paper is trying to accomplish to get into the details of the treatment of the stratosphere in CMAQ. There are papers however, both published and in development, that discuss stratospheric treatment in CMAQ in terms of hemispheric CMAQ model simulations. Those papers do take advantage of some of the measurement data referred to by the reviewer. In short however, there are checks within the CMAQ model to deal with stratospheric ozone, utilizing climatological ozone profiles and comparing those against model simulated upper-level ozone values for consistency. If the values are found to be inconsistent, then the user is warned and in some cases the model simulation is stopped.

C13, Table 2. This table is useful but not easy to read. I suggest that it is converted into a table with several columns:

Simulation name	CMAQ version	WRF version	NEI version	Photolysis scheme	Chemical scheme	Simulation period
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CMAQ_5.0.2_Ba

se

....

Response: The layout of Table 2 has been updated accordingly.

C14, p. 9, l. 4: There are many ways to simulate plume-rise. The scheme that is being used and the underlying data (chimney height, flow speed and temperature if relevant etc.) should be described, or the reader should be referred to a previous publication describing the plume-rise strategy in CMAQ, to permit reproducibility.

Response: The CMAQ plume rise has been described in previous publications (e.g. Foley et al.) and follows the same implementation as SMOKE. A link was added that provides details on the

plume rise calculation used in CMAQ.

Section 4

C15, p. 9, l. 24-30: I think the effect of the changes in the treatment of clouds should also be considered at that point. In July, the increase in O₃ between both versions of WRF is strong in the SW United States (from Louisiana to Virginia). If one goes back to Fig. 1, we can see that this area was represented as cloudy by the CMAQv5.0.2 version with WRF 3.4, but almost cloud-free by WRF 3.7. It appears to me that this reduction of cloudiness between WRF 3.4 and WRF 3.7 is also a very plausible explication for O₃ increase in summertime over this area, particularly as the same is observed in western Mexico and the states of Arizona, Colorado, Utah and New-Mexico.

Response: Agreed that the effect of the change in clouds between the two versions of the model is an important driver in the difference in ozone between the two simulations. The effect of the change in clouds is specifically addressed in section 4.3 that compares the different cloud treatments used in CMAQv5.0.2 and CMAQv5.1. Inherent in this discussion is the effect that the reduced cloudiness in v5.1 has on the ozone mixing ratios.

Section 5

C15, p.13, l. 1-5: I think the word “stochastic” is not appropriate. I very much prefer “subgrid variations”, which is also used. It is very much a modeller vision to consider that everything below grid resolution is essentially stochastic, by the fact that a station close to a highway measures higher contamination levels than a station 3 miles away in a forest is perfectly deterministic.

Response: The term stochastic has been widely used and applied to these subgrid variations, and is the terminology used in the referenced article. We’re just being consistent with the terminology used in that article.

C16, p. 13, l. 6-20: This part describes the increments between both model versions, and would probably be more at its place in Section 4 than in Section 5. section 5 is about the validation of v5.1 so it is confusing that v5.0.2 is mentioned so much at this point.

Response: Section 4 is intended to only focus on a few of the major updates and the impact those specific updates had on the model performance. Section 5 is intended to show the overall performance of the model for ozone and PM_{2.5}. While it would be possible to only show the results of the v5.1 simulation here, it seems useful to also include the v5.0.2 results alongside the v5.1 results so that the reader can quickly identify how they might expect ozone and PM_{2.5} to change between a v5.0.2 and v5.1 simulation. For that reason, we opted to keep results from both model simulations in this section. This is a fairly common way to present results of a model update such as this.

C17, p. 13, l. 15-20: This is a significant caveat that should be mentioned in the model description much earlier than that. The fact that windblown dust treatment was not available for the article simulations and neither for the public release is not anecdotic in my opinion. Also, it would be appreciated that the statement that dust contributions are “small and episodic” is made more quantitative, for example by providing a map of average dust concentrations (and variability) in v5.0.2.

Response: Granted the effect of windblown dust can be large in isolated areas, generally a short amount of time. In the United States, windblown dust concentrations are maximized in the springtime in the desert southwest. We’ve added quantitative values of the springtime seasonal average concentration of soil from CMAQv5.0.2. Overall the seasonal average concentration is

very small compared to the overall PM_{2.5} seasonal average concentration.

C18, Section 5.1:

This section essentially describes the differences between v5.1 and v5.0.2 . While this is pertinent for the study, it does not seem to fit within Section 5, which is about evaluation of v5.1 . Only the parts referring to Fig. 9 and to Figs. S2-S5, such as p. 14, l. 3-14, l. 26,27, etc. partly treat of the evaluation of v5.1. In my opinion, the parts describing differences between v5.1 and v5.0.2 should be moved to Section 4.

Response: This section was intended to serve two purposes. First, it is intended to show the performance of CMAQv5.1 versus observations, which of course is useful to users. Secondly, it is also intended to show the change in performance of ozone and PM_{2.5} between CMAQv5.0.2 and CMAQv5.1 so that users can assess for themselves whether the change in model performance is significant for their application or not. To accomplish this, we present the evaluation results against observations for both versions of the model. Section 4 was intended to only show the impact of the individual model updates, while Section 5 shows the performance of the entire CMAQv5.0.2 and CMAQv5.1 modeling systems. Hopefully the reviewer can agree that this is an acceptable way to accomplish these goals.

C19: figs S2 to S5 should be moved into the main manuscript (because **they do present material permitting an objective evaluation towards the observations in absolute terms, which is lacking in most of the manuscript**). These figures (S2 to S5) also show a rather spectacular improvement from v5.0.2 to v5.1, except maybe for springtime. I think it would be fair that the authors insist more on this very strong improvement of their model results. There are some formatting problems in these Figs. S2-S5 : the simulation name in the caption do not fit exactly the ones given in Tab. 2 , legend of the vertical axis of the top middle panels fail to state that it is the bias which is plotted. If it takes too much space to bring all figures S2-S5 into the manuscript, the authors should maybe consider providing a table with the average observed and modelled values for PM_{2.5} as well as the RMSE and correlations for both model versions, and for the four seasons.

Response: Ideally we would like to include all the figures in the main text, as they all present relevant material. However, it would create an extremely lengthy article with all those figures from the supplemental material included. As a compromise, three new figures have been added that include the observed and modeled seasonal diurnal profiles for PM_{2.5}, O₃ and NO_x. These figures do not include the MB, RMSE and correlation, and the reader is referred to the supplemental figures for that information.

C20, Section 5.2:

I would make essentially the same general recommendations than for Section 5.1: that the parts treating of increments from v5.0.2 to v5.1 be moved into Section 4, and that more focus is put on figures S7-S14, bringing some of them into the manuscript, and/or providing a table with the most relevant statistical parameters for both ozone and NO_x. The title of the section should probably include NO_x as well as ozone.

Response: See responses to C18 and C19.

C21, p. 16, l. 6-12: This difference in summertime ozone concentrations over the eastern US is rather significant and in my opinion can be attributed to the change in meteorology between WRF 3.4 to 3.7 (Fig. 2b): the similarity between Fig. 2b and 10c is striking and the numbers and patterns correspond quite well. I think the authors should comment that, and also the fact that the model bias for ozone in summertime over these regions is increased in v5.1, corresponding to the fact that cloud cover is underestimated in these regions in v5.1 (Fig. 1).

Response: Added to the text the strong correlation between the overall change in ozone in v5.1 and the change in ozone due to the WRF/CMAQ meteorological updates, along with the increase due to increased photolysis in v5.1.

Section 5.2 (“comparison to aircraft measurements”)

C22-1: A general comment about this part is that comparing with a single vertical profile is not enough to state an improvement or a deterioration in a model’s performance. The analysis of other profiles should be included, either from the same campaign, or from routine MOZAIC measurements, which are abundant above the continental US.

Response: The authors agree that titling this section as a comparison to aircraft measurements is a bit misleading even though it does include a comparison to a single day of aircraft measurements. Since the objective of this section was to evaluate the change in model performance for NOY, AN and PNs, the section has been retitled to reflect its purpose. In addition, several statistical metrics of NOY performance have also now been included in the section to help expand the analysis provided and highlight the greatly improved performance of NOY in CMAQv5.1. While it would be nice to be able to show additional profiles from other days and include measurements from other networks, the point of the section was to simply inform the reader of the large improvement in NOY performance and an example of the change in ANs and PNs mixing ratios that can be expected in the new model.

C22 Note, there is a problem in numbering, because previous section is numbered 5.2 as well. I find it very interesting to give some comparison with aircraft measurements, even though it would be great to have it at upper altitude as well, either from this measurement campaign, or from the routine MOZAIC (or equivalent) measurements.

Response: The section number has been corrected.

C23 Please provide the coordinates and altitude of “Edgewood, MD”, as well as the hour (and duration, if relevant) of the considered flight, because PBL structure and the behavior of the real and modelled atmosphere depends a lot on the time of day, and if possible more meteorological context (was it a clear-sky, cloudy, rainy day at that place?). This would help a lot the reader to analyze the figures.

Response: The location and elevation of the Edgewood site have been added to the text. The profiles themselves represent an average of several vertical spirals that took place over Edgewood that day, roughly taking place in the morning, early afternoon and late afternoon. This has now been stated in the text.

C24 Fig. 13: please increase the size of fonts in the panels, it is hard to read in printed version.

Response: The font size has been increased in the figure.

C25 Do the authors have an idea why the Nox (and Noy) values in Fig. 13 are reduced so drastically between both model versions (about 50% for Nox)? Model simulations describe a rather young air mass with Nox/Noy ratio around 60%, while the Nox to Noy ratio in the measurement is about 30%, typical of a much more aged (and clean) air mass, suggesting different trajectories in the model than in reality. Nox level being very dependent on anthropogenic emissions, is it possible that this drastic reduction is due at least in part to the emission update? These changes between model versions seem more dramatic than the smooth statistical changes that appear in the statistical scores between v. 5.0.2. and v. 5.1.

Response: The NO_x and NO_y mixing ratios decrease due to the changes made to the atmospheric chemistry in CMAQv5.1 (Section 2.4.1), while the NO_x mixing ratios would be expected to decrease due to greater photolysis in v5.1 (Section 2.3). On average, the NO_y mixing ratios in CMAQv5.1 decreased 21% in July and 13% in January. So, in the plots shown in Figure 13, the NO_y mixing ratio decrease of about 30% seems reasonable for an isolated case. The emission platform update had little to no impact on the NO_x concentrations, so the emission change does not contribute to the decrease in NO_x seen in the figure.

Section 6

C26 p. 17, l. 21 : these notions are not necessarily familiar to the reader. I think it should be precised that these notions apply to the United States of America, and possibly add a reference that explains what are the SIPs and “Federal rules”.

Response: Added text to explain that SIPs and Federal rules aim to reduce emissions through regulations in order to meet mandated air quality standards.

C27 p. 18, l. 2 : Text and figure caption of Fig. 14 announce that a ratio (emission cut simulation / base simulation) will be shown, but the panels show that what is shown is a “RRF”, a notion which is not defined. If RRF is to be actually used, then it should be defined, and possibly, some clues shall be given about the use of this indicator, which is not known to the entire modelling community.

Response: Since the value presented is indeed a ratio and not a true RRF calculation, the term RRF has been removed from the plots and caption, and replaced with the explanation that a ratio of concentration has been used.

C28 Figure 14: it is not clear to me what kind of samples populates the box plots. Are the samples made from model grid cells, model time series at given locations? Also, the sample size for of each bin should be precised (for example, the appearance of the rightmost box plot for the case of January suggests that the sample size may be very small. A bit more methodological precisions for this plot (as well as Fig. 15) would be welcome. I would also suggest that the format of Fig. 15 is applied to Fig. 14 as well, which avoids introducing the RRF, and permits the reader to evaluate the reduction obtained in v5.0.2, the reduction obtained in v5.1, and visualize and evaluate the difference between the responsiveness in both versions. I think Fig. 14 does not allow as to know if the difference in responsiveness between both model versions amounts to 2% or 50% of the expected model response, while Fig. 15 does.

Response: Figure 14 has been updated to remove the term RRF to the correct description as a daily ratio (as is presented in Figure 15). The bins are populated from model grid cells, which is now stated in the caption. In addition, the number of model grid cells in each bin is now included above the x-axis.

Section 7

C29 p. 18, l. 35: I do not agree that the model has been “evaluated in terms of operational performance” since the evaluation has been performed for year 2011, so more like a reanalysis than an operational forecast model. I suggest to replace by “evaluated by comparison of a simulation of year 2011 to routine measurements of ozone, Nox and PM25 from xxx ground stations” (or something equivalent)

Response: Removed the word “operational” as it is a source of confusion and requires additional context. Here we use the term operational to refer to evaluation against observations, not an

evaluation of the model in an operational (e.g. forecast) mode. However, it seemed easier to remove the word operational and let the evaluation results speak for themselves.

C30 p. 19, l. 10: “to decrease the amount of sub-grid in the photolysis calculation” : please clarify, come words seem to be missing here.

Response: Added the word “clouds” after sub-grid.

C31 p. 19, l. 10-13: it also seems that switching from WRF3.4 to WRF3.7 had a strong effect in reducing model cloudiness over the continental US (Fig. 1), in turn increasing summertime ozone levels over the concerned areas (Fig. 2b), even in the absence of update in the photolysis scheme. Therefore, I find this part of the conclusion (from “The net effect...” to “on average”) a bit questionable.

Response: Hopefully with the addition of the word “clouds” from the above comment it’s clear that we are referring to the decrease in cloudiness in the model as the driving factor in increasing summertime ozone. It’s actually the updates to the cloud treatment in CMAQ (to make them consistent with WRF clouds) and not the transition from WRF3.4 to WRF3.7 that drive the difference in the clouds in CMAQ.

C32 p. 19, l. 13-14: if I am not wrong, these options are not really been described in the main development, neither which one of these options was chosen to obtain the results described here.

Response: The availability of these options was mentioned briefly in Section 2.3 and is mentioned again briefly here as new option available in CMAQv5.1.

C33 p. 20, l. 2: I think the authors should state explicitly which known issues they are referring to (because this may be of interest to model users)

Response: Reworded to remove the words “known issues”. The known issues being referred to was the windblown dust treatment in v5.1, which is referred to in the next sentence anyway.

C34 p. 20, l. 10-11: I think the WRF website should be referred to as well since extensive use of WRF has been made and it seems critical that users use CMAQ with a recent WRF version.

Response: The WRF website was added to the Data availability section.

Overview ~~Description~~ -and evaluation of the Community Multiscale Air Quality (CMAQ) ~~modeling system~~ version 5.1

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Abstract. The Community Multiscale Air Quality (CMAQ) model is a comprehensive multi-pollutant air quality modeling system developed and maintained by the U.S. Environmental Protection Agency's (EPA) Office of Research and Development (ORD). Recently, version 5.1 of the CMAQ model (v5.1) was released to the public, ~~which~~ incorporatinges a large number of science updates and extended capabilities over the previous release version of the model (v5.0.2). These updates include improvements in the meteorological calculations in both CMAQ and the Weather Research and Forecast (WRF) model used to provide meteorological fields to CMAQ; updates to the gas and aerosol chemistry; revisions to the calculations of clouds and photolysis; and improvements to the dry and wet deposition in the model. Sensitivity simulations isolating several of the major updates to the modeling system show that changes to the meteorological calculations generally result in greater~~enhanced~~ afternoon and early evening mixing in the model, times-periods when the model historically underestimates mixing. This enhanced mixing e-results is higher ozone (O₃) mixing ratios on average due to reduced NO titration, and lower fine particulate matter (PM_{2.5}) concentrations due to greater dilution of primary pollutants (e.g. elemental and organic carbon). Updates to the clouds and photolysis calculations greatly ~~greatly~~ improve consistency between the WRF and CMAQ models and result in generally higher O₃ mixing ratios, primarily due to reduced cloudiness and ~~reduced~~ attenuation of photolysis in the model. Updates to the aerosol chemistry results in higher secondary organic aerosol (SOA) concentrations in the summer, thereby reducing summertime PM_{2.5} bias (PM_{2.5} is typically underestimated by CMAQ in the summer), while updates to the gas chemistry results in generally-slightly increased~~higher~~ O₃ and PM_{2.5} on average in January and July ~~(small) and slightly higher PM_{2.5} concentrations on average in both January and July.~~ Overall, the seasonal variation in simulated PM_{2.5} generally improves in the new model version CMAQv5.1 (when considering all model updates), as PM_{2.5} concentrations decrease in the winter (when PM_{2.5} is generally overestimated ~~by CMAQ v5.0.2~~) and increase in the summer (when PM_{2.5} is generally underestimated ~~by CMAQ v5.0.2~~). Ozone mixing ratios are higher on average with v5.1 versus v5.0.2, resulting in higher O₃ mean bias, as O₃ tends to be overestimated by CMAQ throughout most of the year (especially at locations where the observed O₃ is low), however O₃ both the error and correlation are~~is~~ largely improved with v5.1. Sensitivity simulations for several hypothetical emission reduction scenarios showed that v5.1 tends to be slightly more responsive to reductions in NO_x (NO + NO₂), VOC and SO_x (SO₂ + SO₄) emissions than v5.0.2, representing an improvement as previous studies have shown CMAQ to underestimate the observed reduction in O₃ due to large, widespread reductions in observed emissions. ~~Finally, the computational efficiency of the model was significantly improved in v5.1, which keeps runtimes similar to v5.0.2 despite the added complexity to the model.~~

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Keywords

CMAQ, WRF, Air Quality Modeling, Model Evaluation, Ozone, Particulate Matter, PM_{2.5}

1 Introduction

Numerous Federal (e.g. United States Environmental Protection Agency (USEPA)), State and private entities rely on numerical model simulations of atmospheric chemistry, transport and deposition of airborne emissions and the resulting pollutants as part of their decision-making process for air quality management and mitigation (e.g. Scheffe et al., 2007). Chemical Transport Models (CTMs), such as the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006), are often employed to provide information about the potential effects of emission control strategies (e.g. Fann et al., 2009), climate change (e.g. Nolte et al., 2008), and provide next-day air quality forecasts (e.g. Eder et al., 2006) in order to inform and protect the public from potentially harmful air pollutants. Since these models are often used to inform the standard setting and implementation for criteria pollutants (e.g. ozone (O₃) and fine particulate matter (PM_{2.5})), they must be maintained at the state-of-the-science. New versions of the CMAQ model have been released periodically over the past fifteen years, with each new version consisting of numerous updates to the scientific algorithms within the model, while also improving the quality of the input data used. Collectively, these updates are aimed at improving the underlying science of atmospheric dynamics and chemistry represented in the model, extending the capabilities for emerging applications, and reducing systematic biases in the modeling system. Every new release of the CMAQ model undergoes extensive evaluation in order to establish its credibility (e.g. Mebust et al., 2003; Appel et al., 2007, 2008, 2013; Foley et al., 2010) and documents its performance relative to previous versions. Most recently, the CMAQ modeling system version 5.1 (v5.1) has been tested and evaluated against observations and was publically released in December 2015 (<http://www.cmaq-model.org/>).

The scientific upgrades in the CMAQv5.1 modeling system include major revisions to the Pleim-Xiu land-surface model (PX-LSM; Pleim and Xiu, 1995) and the Asymmetric Convective Mixing version 2 (ACM2; Pleim, 2007ab) planetary boundary layer (PBL) model in the Weather Research and Forecast (WRF) model version 3.7 (Skamarock et al., 2008), which required revisions to the ACM2 scheme in CMAQ to maintain consistency. Corrections were also made to the Monin-Obukhov length (MOL) calculation in CMAQv5.1 to make it consistent with the calculation in the WRF model. The changes to the PX-LSM, ACM2 and MOL calculations in CMAQ had significant impact on the mixing within both WRF and CMAQ, and hence large impacts on the pollutant concentrations in CMAQ. These updates are described in Section 2.1. A new explicit treatment of secondary organic aerosol (SOA) formation from isoprene, alkenes and PAHs was also added in CMAQv5.1. Additionally, two aerosol mechanisms are now available in v5.1, AERO6 and AERO6i (with isoprene extensions), which include updates to the SOA and ISORROPIA algorithms (Nenes et al., 1998; Nenes et al., 1999). The AERO5 mechanism has been deprecated and is no longer available. The updates to the aerosol treatment in v5.1 are described in Section 2.2. Significant changes were also made to the in-line calculation of photolysis rates (described in Section 2.3). Finally, the changes to the photochemistry in v5.1 underwent major changes, specifically new explicit treatment of secondary organic aerosol (SOA) formation from isoprene, alkenes and PAHs; updates to the photochemical cross sections and quantum yields for the Carbon Bond 2005 e51 (CB05e51) chemical mechanism were updated; along with updates to inorganic and organic chemical reaction rates and products to ensure consistency with the International Union of Pure and Applied Chemistry (IUPAC); and, And finally the additional representation of organic nitrate species in CB05e51. Additionally, two aerosol mechanisms are now available in v5.1, AERO6 and AERO6i (with isoprene extensions), which include updates to the SOA and ISORROPIA algorithms (Nenes et al., 1998; Nenes et al., 1999), while AERO5

has been deprecated and is no longer available. Significant changes were also made to the in-line calculation of photolysis rates. Additionally, two aerosol mechanisms are now available in v5.1, AERO6 and AERO6i (with isoprene extensions), which include updates to the SOA and ISORROPIA algorithms (Nenes et al., 1998; Nenes et al., 1999), while AERO5 has been deprecated and is no longer available. There were also major revisions to the Pleim-Xiu land surface model (PX-LSM; Pleim and Xiu, 1995) and Asymmetric Convective Mixing version 2 (ACM2; Pleim, 2007ab) planetary boundary layer (PBL) model in the Weather Research and Forecast (WRF) model version 3.7 (Skamarock et al., 2008), which required revisions to the ACM2 scheme in CMAQ to maintain consistency. In addition, corrections were also made to the Monin-Obukhov length (MOL) calculation in v5.1 to make it consistent with the calculation in the WRF model. The changes to the PX-LSM, ACM2 and MOL calculations in CMAQ had significant impact on the mixing within both WRF and CMAQ, and hence large impacts on the pollutant concentrations in CMAQ. These updates are described in Section 2.4.

Section 2 provides a brief description of the major scientific and structural improvements included in v5.1. The model configuration and observational data sets used in the model evaluation are provided in Section 3. The evaluation of v5.1 itself is then presented in two parts. Section 4 documents the evaluation of several specific changes that were isolated as part of the overall testing of the model. Specifically, (a) Section 4.1 evaluates the changes to the PX-LSM and ACM2 schemes meteorological updates in WRF and CMAQ; Section 4.2 evaluates the (b) updates to the MOL calculation aerosol updates; Section 4.3 evaluates the (c) changes to the inline photolysis calculation and the representation of clouds within CMAQv5.1; and (d) Section 4.4 evaluates the updates to the CB05e51 chemical mechanism. These increments were chosen as the focus of this paper because they represent a fundamental change from the previously released model version and had the propensity to impact model performance for criteria pollutants. The second portion of the evaluation, presented in Section 5, summarizes the overall change in PM_{2.5} and O₃ model performance with v5.1 compared to the previously released version (CMAQ version 5.0.2 (v5.0.2)). Section 6 provides a discussion of the model response of O₃ and PM_{2.5} to hypothetical reductions in emissions. And finally Section 7 discusses upcoming changes and future work for CMAQv5.2 (v5.2), followed by a summary discussion in provided in Section 8.

2 Review of scientific improvements in CMAQ v5.1

Improvements to the v5.1 modeling system are the result of many years of scientific advancements derived from laboratory, field and numerical experiments and the efforts of a relatively small group of model developers that both investigate avenues for model improvements and then update the model (i.e. write code). Given the large community of CMAQ model users, there are never sufficient resources to diagnose and address every issue in the modeling system that has been reported. As such, it is necessary to prioritize updates to the model based on many different factors, including results from evaluations of past model versions, existing and upcoming regulatory needs, emerging scientific issues, requests from the CMAQ user community, and the expertise within the model developer group to meet those needs/requests. The updates described herein represent the “major” updates made to the CMAQ modeling system from the previous model version, and therefore does not constitute a fully comprehensive description of all the changes made to the system. This section briefly describes these “major” updates to CMAQ, providing the reader with an understanding of what was updated in the model and why. A comprehensive description of all the updates made in v5.1 and in-depth technical documentation of those changes can be found on the CMAS Center website for the CMAQ v5.1 release at [https://cmaswiki-cempd.vipapps.unc.edu/index.php/CMAQ_version_5.1_\(November_2015_release\)_Technical_Documentation](https://cmaswiki-cempd.vipapps.unc.edu/index.php/CMAQ_version_5.1_(November_2015_release)_Technical_Documentation).

2.1 WRF and CMAQ meteorological and transport updates

The WRF and CMAQ models were updated to improve the representation of land-surface processes and vertical mixing. There were two changes made to the PX-LSM in WRF. First, the stomatal conductance function for photosynthetically active radiation (PAR) was revised based on measurements of net photosynthetic rate as a function of PAR for cotton plants reported by Echer and Rosolem (2015). The new functions yield a significantly lower magnitude when short-wave radiation is less than 350 Wm⁻². This in turn results in reduced latent heat flux and enhanced sensible heat flux, causing a delay in surface stabilization (prolongs mixing) during evening transitions hours (i.e. sunset). This reduces overestimations (reduced positive bias) in water vapor mixing ratios, which are common in the WRF-CMAQ modeling system during the evening transition. Similarly, overestimation of concentrations of surface emitted species (e.g. NO, NO₂, CO and EC) are also reduced during the evening transition. This change was released in WRFv3.7 and further revised in WRFv3.8. The second change made to the PX-LSM is an increase of the coefficient to the surface energy forcing in the soil temperature force-restore equation (C_v), which is related to volumetric heat capacity (c_v) and heat conductivity (λ) (Pleim and Gilliam 2009) as:

$$C_v = 2 \left(\frac{\pi}{c_v \lambda \tau} \right)^{1/2}$$

where τ is 1 day (86400 s), from the previous value of $8 \times 10^{-6} \text{ K m}^2 \text{ J}^{-1}$ recommended by Giard and Bazile (2000) to $1.2 \times 10^{-5} \text{ K m}^2 \text{ J}^{-1}$. The new value for C_v results from updated values for c_v and λ for vegetation based on measurements of various leaves by Jayakshmy and Philip (2010) ($c_v = 2.0 \times 10^6 \text{ J m}^{-3} \text{ K}^{-1}$, $\lambda = 0.5 \text{ W m}^{-1} \text{ K}^{-1}$). These changes reduce overestimations of minimum 2-meter temperature (i.e. warmer surface temperatures) during the early morning (dawn) hours while also reducing underestimations of 2-meter temperature during the post-dawn hours. The WRF and CMAQ models were updated to improve the representation of land-surface processes and vertical mixing. There were two changes made to the PX-LSM in WRF. First, the stomatal conductance function for photosynthetically active radiation (PAR) was revised with a new function that yields a significantly lower magnitude when short-wave radiation is less than 350 Wm⁻². This in turn results in reduced latent heat flux and enhanced sensible heat flux, causing a delay in surface stabilization (prolongs mixing) during evening transitions hours (i.e. sunset). This reduces overestimations (reduced positive bias) in water vapor mixing ratios, which are common during the evening transition. Similarly, overestimation of concentrations of surface emitted species (e.g. NO, NO₂, CO and EC) are also reduced during the evening transition. This change was released in WRFv3.7 and further revised in WRFv3.8. The second change made to the PX-LSM is a reduction in the heat capacity value for vegetation based on values suggested by Jayakshmy and Philip (2010), which reduces overestimations of minimum 2-meter temperature (i.e. warmer surface temperatures) during the early morning (dawn) hours while also reducing underestimations of 2-meter temperature during the post-dawn hours.

There were also two major revisions made to the ACM2 vertical mixing scheme in both WRF and CMAQ. In WRF, the ACM2 was updated to estimate and apply different eddy diffusivities for momentum (K_m) and heat (K_h) so that the Prandtl number (Pr) is no longer assumed to be unity ($Pr = K_m/K_h \neq 1$). The second major modification to ACM2 is the implementation of new stability functions for both heat and momentum for stable conditions, which allows for more mixing in the stable regimes, particularly moderately stable conditions that often occur in the early evening hours. CMAQ v5.1 has also been modified to include the same stability functions that are used in WRF v3.7, and therefore, for consistency, WRF v3.7 (or newer) and CMAQ v5.1 should be used together. Both of these revisions to the ACM2 are described in Pleim et al. (2016). There were also two major revisions made to the ACM2 vertical mixing scheme in both WRF and CMAQ (Pleim et al., 2016). In WRF, the ACM2 was updated to estimate and apply different eddy diffusivities for momentum (K_m) and heat (K_h) so that the Prandtl number (Pr) is no longer assumed to

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be unity ($Pr = K_m/K_h \neq 1$). The second major modification to ACM2 is the implementation of new stability functions for both heat and momentum for stable conditions, which allows for more mixing in the stable regimes, particularly moderately stable conditions that often occur in the early evening hours. CMAQ v5.1 has also been modified to include the same stability functions that are used in WRF v3.7, and therefore, for consistency, WRF v3.7 (or newer) and CMAQ v5.1 should be used together.

The Monin-Obukhov length (MOL) values used in the ACM2 model in CMAQ were found to differ from the MOL values used in the ACM2 model in WRF. Specifically, the output from WRF was for a preliminary estimate of MOL that was computed in the surface layer model in WRF (module_sf_pxsfc_lay.F). The MOL was later re-computed in ACM2 in WRF but not loaded into the output array. This inconsistency has been fixed in v5.1 by re-computing the MOL in CMAQ exactly as it is computed in ACM2 in WRF. However, starting with WRF v3.8, this re-computed MOL value will be available in the WRF output, and therefore it will be unnecessary to re-compute the MOL value in CMAQ. ~~In addition, the estimated MOL value in the surface layer model in WRF will be improved such that there is little difference between the initial MOL estimate and the final re-calculated value.~~

~~Finally, previous evaluations of the ground-level coarse-particle (PM_{10}) concentrations in CMAQ have shown that the model significantly underestimated the total PM_{10} concentrations (Appel et al., 2012). Contributing to this underestimation is the fact that CMAQ did not have a mechanism in place to allow coarse particles to settle from upper layers to lower layers (although coarse particles in layer one can settle to the surface). As a result, large particles that would normally settle to the lower layers could remain trapped in the layers in the model in which they are emitted or formed. To account for this deficiency in the model, the effects of gravitational settling of coarse aerosols from upper to lower layers has been added to v5.1 to more realistically simulate the aerosol mass distribution. The net effect of this update is an increase in ground-level PM_{10} concentrations in v5.1 compared to v5.0.2, particularly near coastal areas impacted by sea spray (Nolte et al., 2015).~~

2.2 Scientific improvements in the CMAQ v5.1 aerosol treatment

CMAQ has historically underestimated SOA in both urban (Woody et al., 2016) and rural (Pye et al., 2015) locations. Thus, improvements to the representation of aerosol from anthropogenic and biogenic hydrocarbons were needed. The updates to SOA formed from anthropogenic volatile organic compounds (VOC) focus on VOC compounds in existing emission inventories, such as the EPA National Emissions Inventory (NEI), that are likely to fall in the intermediate VOC (IVOC) range. These include long-chain alkanes such as heptadecane and polycyclic aromatic hydrocarbons (PAHs) such as naphthalene. Since these compounds are much less volatile than traditional VOCs, they readily form aerosol in high yields. Long-chain alkanes and PAHs were included in other VOC categories in CMAQ versions prior to v5.1, but were lumped with smaller, more-volatile compounds that did not form SOA with the same efficiency. By separating long-chain alkanes and naphthalene at the emission processing step, CMAQ can better account for their higher yields. Several studies (e.g. Pye and Pouliot, 2012; Jathar et al., 2014) Work by Pye and Pouliot (2012) as well as Jathar et al. (2014) indicate have indicated that a large fraction of VOC emissions, particularly IVOC-type compounds, may not be characterized in emission inventories, which limits how much SOA can be formed from anthropogenic VOCs in current chemical transport models.

Several new SOA species were introduced in v5.1 AERO6, specifically AALK1 and AALK2 (from long-chain alkanes) and APAH1, APAH2, and APAH3 (from naphthalene). CMAQ v5.1 predicted alkane SOA is responsible for ~20 to 50% of SOA from anthropogenic VOCs, with the largest absolute concentrations occurring during summer in urban areas. Naphthalene oxidation is predicted to produce more modest amounts of SOA (Pye and Pouliot, 2012). Note that PAH SOA in v5.1 only considers

naphthalene as the parent hydrocarbon, which about half of the PAHs is considered as SOA precursors in Pye and Pouliot (2012). This approach was used since naphthalene is a high priority hazardous air pollutant (HAP) and necessary in the model for purposes other than SOA formation.

5 CMAQv5.1 includes has been updated to updates to present the include the isoprene epoxydiols (IEPOX) SOA resulting from aqueous reactions for most chemical mechanisms including CB05 and SAPRC07 as described in Pye et al. (2013). Later generation isoprene oxidation products formed under low-NO_x conditions, specifically ~~isoprene epoxydiols (IEPOX)~~, are recognized as a significant source of SOA based on laboratory (Surratt et al. 2010), field (Hu et al. 2015), and modeling (McNeill et al. 2012, Pye et al. 2013, Marais et al. 2016) studies. This SOA is linked to sulfate and acidity and thus represents an anthropogenically controlled source of biogenic SOA. CMAQv5.1 includes updates to present the IEPOX SOA resulting from aqueous reactions for most chemical mechanisms including CB05 and SAPRC07 as described in Pye et al. (2013).

In addition to the SOA updates for anthropogenic VOCs, AISO₃ (acid catalyzed isoprene epoxide aerosol) was also revised in CMAQv5.1 to represent SOA from IEPOX. For the CB05tuc1, CB05e51 and SAPRC07 chemical mechanisms with IEPOX formation in the gas-phase, heterogeneous uptake of IEPOX on acidic aerosol results in SOA (Pye et al., 2013). This IEPOX SOA replaces the AISO₃ treatment based on Carlton et al. (2010). The AISO3J species name is now retained for IEPOX SOA and represents the sum of IEPOX-derived organosulfates and 2-methyltetrols. Explicit isoprene SOA species including 2-methyltetrols, 2-methylglyceric acid, organosulfates, and oligomers/dimers are available in the SAPRC07tic with AERO6i mechanism now available in CMAQv5.1. See Table 1 for more information regarding these new SOA species.

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2.3 Improvements to the CMAQv5.1 in-line photolysis and cloud model

25 The in-line calculation of photolysis rates in CMAQ has undergone significant changes. The calculation of photolysis rates in v5.1 still uses the same approach for calculating actinic fluxes by solving a two-stream approximation of the radiative transfer equation (Binkowski et al., 2007; Toon et al., 1989) over wavebands based on the FAST-J photolysis model (Wild et al., 2000). Each layer includes scattering and extinction using simulated air density, cloud condensates, aerosols, and trace gaseous such as O₃ and NO₂. The first area changed in v5.1 is how clouds are described in the actinic flux calculation. In v5.0.2, a vertical column had a single cloud deck with constant cloud fraction, liquid water content and water droplets as the source of scattering and extinction from clouds. These cloud parameters were diagnosed from humidity and air temperature predicted by the meteorological model (e.g WRF). CMAQv5.1 uses additional information available from WRF that describes the resolved cloud cover, which allows the vertical column to have multiple cloud decks with variable cloud fractions and multiple types of water condensates. In addition to the resolved cloud cover, v5.1 also includes the radiative effect from CMAQ's sub-grid convective clouds in the calculation of actinic fluxes. CMAQ uses the ACM cloud model to describe sub-grid convective clouds based on convective precipitation rates from WRF. These updates to the clouds used in the photolysis rates improved CMAQ's internal consistency between cloud mixing, aqueous chemistry, and gas-phase chemistry.

35 The second area of change to the in-line photolysis calculation addressed the radiative effect from aerosols. The mixing model used to compute the refractive indices of aerosol modes (an internal-volume weighted average model) allows the refractive index of each aerosol component to depend on wavelength. Most importantly, the refractive index for elemental (black) carbon reflects the current scientific consensus (Bond and Bergstrom, 2006; Chang and Charalampopoulos, 1990; Segelstein, 1981; Hess et al.,

1998) and increases its absorptive capacity from the value v5.0.2. Additionally, estimating aerosol optical properties includes new options to solve Mie scattering theory, or the option to use the Core-Shell model with an elemental carbon core (Bohren and Huffman, 2004). A user can choose to use these options by setting environment variables before executing the CMAQ model (http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQv5.1_In-line_Calculation_of_Photo%20lysis_Rates). By default, v5.1 uses approximate solutions to Mie scattering and the internal-volume weighted average model (Binkowski et al., 2007). Third, several new variables (e.g. resolved cloud fraction, sub-grid cloud fraction, resolved cloud water content) have been added to the cloud diagnostic file that describe the optical properties of aerosol and clouds and their radiative effects. The in-line calculation of photolysis rates has undergone significant changes in three areas. First, the description of clouds has changed. In v5.0.2, a vertical column had a single cloud deck with a constant cloud fraction and water droplet mixing ratio. In v5.1, a vertical column can have multiple cloud decks with variable cloud fractions and multiple types of water condensates. The new description is more consistent with the WRF meteorological model output typically used for CMAQ simulations. Second, the mixing model used to compute the refractive indices of aerosol modes, an internal-volume weighted average model, allows the refractive index of each aerosol component to depend on wavelength. Most importantly, the refractive index for elemental (black) carbon reflects the current scientific consensus (Bond and Bergstrom, 2006; Chang and Charalampopoulos, 1990; Segelstein, 1981; Hess et al., 1998) and increases its absorptive capacity. Additionally, estimating aerosol optical properties includes new options to solve Mie scattering theory or to use the Core-Shell model with an elemental carbon core (Bohren and Huffman, 2004). Run-time options determine whether to solve Mie scattering or to use the Core-Shell model for the internal mixed aerosol modes (http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQv5.1_In-line_Calculation_of_Photo%20lysis_Rates). By default, the model uses approximate solutions to Mie scattering and the internal-volume weighted average model (Binkowski et al., 2007). Third, several new variables (e.g. resolved cloud fraction, sub-grid cloud fraction, resolved cloud water content) have been added to the cloud diagnostic file that describe the optical properties of aerosol and clouds and their radiative effects.

Cloud albedo from NASA's Geostationary Operational Environmental Satellite Imager product (GOES; <http://satdas.nsstc.nasa.gov/>) was used to evaluate the cloud parameterizations in WRF3.7 and in the photolysis calculations within CMAQ. The GOES product has a 4km horizontal resolution and was re-gridded to the 12 km grid structure used in the WRF and CMAQ simulations using the Spatial Allocator utility (<https://www.emascenter.org/sa-tools/>). The satellite data are available at 15 minutes prior to the top of the hour during daytime hours (11:45UTC—23:45UTC) and were matched to model output at the top of the hour. Figure 1 shows the average cloud albedo during daytime hours in July 2011 derived from (a) the GOES satellite product (b) WRF3.7 (c) CMAQv5.0.2 photolysis calculations (d) CMAQv5.1 photolysis calculations. Comparison of Figure 1(a) to (c) shows that the cloudiness parameterization in the photolysis module in v5.0.2, which was based solely on relative humidity, produced far too many clouds relative to the satellite observations. The new parameterization within v5.1 uses the resolved cloud fractions and water content from WRF and sub-grid cloud fractions and water content determined by the convective cloud model within CMAQ (acm_ae6; Pleim et al., 2005). As a result, the model predicted clouds in v5.1 are now considerably more consistent with the WRF parameterization (compare Figure 1 (b) to (d)). Two notable issues remain with the v5.1 modeled cloud parameterization. The photolysis cloud parameterization in v5.1 produces more clouds over water compared to the WRF parameterization, which is itself biased high for some parts of the Atlantic Ocean compared to GOES. This issue will be addressed by science updates planned for the CMAQ system and evaluation results are expected to improve in the next CMAQ release (See Section 7.4). A more significant issue, from an air quality perspective, is the under prediction of clouds over much of the Eastern and West Central US in the WRF predicted clouds, which is now directly passed along to CMAQ. This misclassification of modeled clear sky conditions can contribute to an over prediction of O₃ in these regions. Resolving this issue will require changes to the WRF cloud parameterization. Future research will also include changing the

sub-grid cloud treatment currently used in the CMAQ system to be consistent with the sub-grid parameterization used in WRF. Section S.1 in the supplemental material provides a table with additional evaluation metrics of the modeled clouds over oceans versus over land and also describes how cloud albedo was calculated for the three model simulations.

2.4 Improvements in CMAQ v5.1 atmospheric chemistry

Several changes were made to the CB05TUCL chemical mechanism in v5.1 (Whitten et al., 2010; Sarwar et al., 2012), which is now referred to as CB05e51. These changes include updates to reactions of oxidized nitrogen (NO_x) species; incorporation of new research on the atmospheric reactivity of isoprene photo-oxidation products; addition of several high priority HAPs to the standard CB05e51 mechanism (following the protocol in the multipollutant version of CMAQ); and other changes to update the mechanism and make it compatible with updates to the aerosol chemistry, but overall retaining the fundamental core of the CB05 mechanism. The objective was to limit modifications to those reactions that are most important, so that the core CB05 mechanism was not fundamentally changed. A more detailed explanation of the changes made in the CB05e51 mechanism is provided below.

2.4.1 NO_x updates and additions

The most extensive changes made consisted of updates and extensions of the NO_x species, including peroxyacetyl nitrates, alkyl nitrates, and NO_x reactions with HO_x . The thermal formation and degradation of peroxyacetyl nitrate (PAN) were modified to correct the parameters that describe the rate constant pressure dependence in the fall-off region between the high-pressure limit and the low-pressure limit (i.e. using $N=1.41$ and $Fe=0.3$ instead of the CB05tuel defaults of $N=1.0$ and $Fe=0.6$) based on the values determined by (Bridier et al., 1991). An additional species, MAPAN, was added to explicitly represent PANs from methacrolein because these are a possible contributor to SOA formation. The $\text{OH}+\text{NO}_2$ reaction rate was updated based on (Tropea, 2012) and a small yield of HNO_3 (<1% at standard temperature and pressureSTP, varying with temperature and pressure) was added to the reaction of HO_2+NO (Butkovskaya et al., 2007). The single alkyl nitrate species (~~NTR~~) in CB05, NTR, was replaced with seven species to better investigate the variety of chemical and physical fates of alkyl nitrates. The first-generation monofunctional alkyl nitrates and difunctional hydroxy nitrates were assigned Henry's law constants of $6.5\text{e-}1$ M and $6.5\text{e}3$ M respectively, while second generation carbonyl nitrates were assigned $1.0\text{e}3$ M and multifunctional hydroxynitrates were assigned a value of $1.7\text{e}4$ M. Five species are predominantly from anthropogenic sources, with the relative distribution of mono-functional (alkyl nitrates) and multi-functional (hydroxy, carbonyl, hydroxycarbonyl, and hydroperoxy) nitrate products determined based on the nitrates produced from the five alkanes and alkenes, with the largest emissions as listed in the NEI (Simon et al., 2010). The other two nitrate species represent first generation and later generation nitrates from biogenic (isoprene and terpene) sources. Biogenic nitrate products were based on reaction products from Lee et al. (2014), with NO_x recycling from secondary biogenic nitrate products (Jenkin et al., 2015) and photolysis rates with quantum yields of unity. Finally, a heterogeneous hydrolysis rate of alkyl nitrates was added (Hildebrandt-Ruiz et al., 2013), with a six-hour lifetime on aerosol at high relative humidity (Liu et al., 2012; Rollins et al., 2013). Additional details can be found in the CMAQv5.1 release documentation ([http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQ_version_5.1_\(November_2015_release\)_Technical_Documentation](http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQ_version_5.1_(November_2015_release)_Technical_Documentation)).

2.4.2 Other changes

The high HO_x pathways for isoprene oxidation have been modified to explicitly account for production of isoprene-epoxydiol (IEPOX), which can form SOA and modify the gas-phase concentrations. The high NO_x pathways have been modified to

explicitly produce methacrolein PAN (MAPAN, described in Section 2.4.1) because it reacts faster with OH than other PAN species. Several high priority HAPs were added to the standard version of CB05e51 as either active species or reactive tracers, specifically acrolein, 1,3-butadiene (which produces acrolein), toluene, xylene isomers, α - and β -pinene, and naphthalene using reaction pathways and rates as defined by IUPAC. Refer to the CMAQv5.1 release documentation for additional details on these updates.

Several other, smaller changes were made to the chemistry to either improve consistency with IUPAC, enhance the integration with heterogeneous chemistry, or for numerical consistency. These include the updates to the products of ethanol reaction with OH using recommended yields from IUPAC (<http://iupac.pole-ether.fr>; accessed May 11, 2016); updates to the reactions of acylperoxy radicals with HO₂ to include a 44% yield of OH; the addition of a new species, SOAALK, to account for SOA formation from alkanes; and the addition of gas-phase and heterogeneous nitryl chloride formation (CINO₂) and CINO₂ photolysis as described by Sarwar et al. (2012).

2.5 Updates to air-surface exchange processes in CMAQ v5.1

Meteorologically dependent emissions and deposition, hereafter referred to as air-surface exchange, were extensively updated in v5.1. A data module was developed to share meteorological and calculated atmospheric transport environmental variables between vertical diffusion, deposition, and meteorological dependent emissions to more consistently represent processes common to both deposition and emissions. Additionally, sea salt and biogenic emissions and dry deposition routines were updated.

2.5.1 Sea salt aerosol emission

The sea salt aerosol emissions module was updated to better reflect emissions estimates from recent field observations and to incorporate ocean thermodynamic impacts on emissions. The size distribution of sea salt aerosol was expanded to better reflect recent fine-scale aerosol measurements in laboratory and field studies (de Leeuw et al., 2011) by modifying the Θ parameter of Gong (2003) from 30 to 8. A sea-surface temperature (SST) dependency to the sea-salt aerosol emissions following Jaeglé et al. (2011) and Ovadnevaite et al. (2014) was also added, which increased accumulation and coarse mode sea-salt emissions in regions with high SSTs and reduced the emissions in regions with low SSTs. Finally, the surf-zone emissions of sea-salt aerosol were reduced by 50% assuming a decrease in the surf-zone width from 50 m to 25 m to address a systematic overestimation of near-shore coarse sea-salt aerosol concentrations (Gantt et al., 2015).

2.5.2 Biogenic emissions (BEIS)

There were also several updates to the calculation of non-methane biogenic volatile organic carbon (BVOC) emissions in v5.1. The Biogenic Emissions Inventory System (BEIS; <https://www.epa.gov/air-emissions-modeling/biogenic-emission-inventory-system-beis>) model was updated to include the implementation of a dynamic two-layer, sun and shaded, vegetation canopy model, while the PAR response function was integrated into the canopy model following Niinemets et al. (2010) for each canopy layer. In earlier versions of BEIS, emissions were a function of the 2-meter temperature which was inconsistent with measured emission factors that were empirically correlated with leaf temperature. BEIS 3.6.1 released with v5.1 was updated to model emissions as a function of the leaf temperature rather than 2-meter temperature to be more consistent with how BVOC emission factors are typically estimated. For additional details see Bash et al. 2016. A leaf temperature algorithm was implemented that replaced the 2-meter temperature to be more consistent with emission factor measurements. Finally, the Biogenic Emission Land-use Data (BELD) version 4.0) and emission factors for herbaceous wetlands were updated to address overestimates of

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biogenic BVOCs at coastal sites (Guenther et al., 2006), and ~~and updated the~~ BELD land-use and vegetation species ~~were updated~~ with using high-resolution satellite data and in-situ survey observations from 2002-2012 (Bash et al., 2016).

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2.5.3 Dry deposition

~~TFinally,~~ there were two important updates to the dry deposition calculation in v5.1. First, the dry deposition of O₃ over oceans was updated to include the additional sink due to interaction with iodide in the seawater (marine halogen chemistry), with the iodide concentrations estimated based on sea-surface temperature (Sarwar et al., 2015), which increased the O₃ deposition velocity over oceans. Second, over vegetative surfaces, the wet cuticular resistance was updated following Altimir et al. (2006), 385 s m⁻¹, and dry cuticular resistance was set to the value of Wesley (1989) for lush vegetation, 2000 s m⁻¹. These changes resulted in an approximately 2.0 ppbv reduction in the modeled O₃ mixing ratios, with the largest reductions, ~10%, occurring during the nighttime and early morning hours, and approximately a 2% reduction in the modeled midday O₃ mixing ratio.

2.5.4 Gravitational Settling

~~Finally,~~ Previous evaluations of the ground-level coarse particle (PM₁₀) concentrations in CMAQ have shown that the model significantly underestimated the total PM₁₀ concentrations (Appel et al., 2012). Contributing to this underestimation is the fact that CMAQ previously did not have a mechanism in place to allow coarse particles to settle from upper layers to lower layers (although coarse particles in layer one can settle to the surface). As a result, large particles that would normally settle to the lower layers in the model could remain trapped in the layers in the model in which they were emitted or formed. To account for this deficiency in the model, the effects of gravitational settling of coarse aerosols from upper to lower layers has been added to v5.1 to more realistically simulate the aerosol mass distribution. The net effect of this update is an increase in ground-level PM₁₀ concentrations in v5.1 compared to v5.0.2, particularly near coastal areas impacted by sea-spray (Nolte et al., 2015).

As stated in the beginning of this section, but is useful to reiterate here, the information provided in this section only covers a portion of the vast number of updates that went into v5.1, and was intended to make the reader aware of the more significant changes made and why, but often avoids including the very specific detailed code changes that were made to the model. Those seeking a complete detailed list of all the changes made to the model should refer to the v5.1 technical documentation using the link provided at the beginning of this section.

3 Modeling setup and observational data sets

The modeling setup for the evaluation of v5.1 utilizes a domain covering the entire contiguous United States (CONUS) and surrounding portions of northern Mexico and southern Canada, and the eastern Pacific and western Atlantic oceans. The modeling domain consists of 299 north-south by 459 east-west grid cells ~~with utilizing~~ 12-km by 12-km horizontal grid spacing and 35 vertical layers with varying thickness extending from the surface to 50 hPa and an approximately 10-meter mid-point for the lowest (surface) model layer. The simulation time period covers the year 2011, which is a base year for the ~~National Emission Inventory (EPA's NEI)~~ and also a period during which specialized measurements from a variety of trace species are available from the Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ; http://www.nasa.gov/mission_pages/discover-aq/index.html) campaign.

All the CMAQ simulations presented here employed the Euler Backward Iterative (EBI) solver. The v5.0.2 simulations utilized the windblown dust treatment available, while the v5.1 simulations did not due to errors in the implementation of the windblown dust model in v5.1. However, the overall contribution of windblown dust to $PM_{2.5}$ is small on a seasonal average and does not affect the seasonal comparisons shown in Section 5. Additional details regarding the options employed in the CMAQ simulations are available upon request from the corresponding author. For the annual simulations, a 10-day spin-up period in December 2010 was used (and then discarded) to reduce the effects of the initial conditions, after which the model was run continuously for the entire year 2011 (one continuous simulation stream). For the one-month January and July sensitivity simulations presented, 10-day spin-up periods in the previous month were used and then discarded. Boundary conditions for the 12-km CMAQ simulations are provided by a 2011 hemispheric GEOS-Chem (Bey et al., 2001) with the chemical species mapped to the corresponding CMAQ species.

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Several sets of CMAQ simulations were performed to help thoroughly evaluate both the overall change in model performance between v5.0.2 and v5.1 and to examine the individual impact of specific model process changes on the model performance metrics. As such, different input data sets were used/required for the v5.0.2 and v5.1 simulations. The base v5.0.2 simulation (CMAQv5.0.2_Base) utilized WRF v3.4 meteorological input data, while WRF v3.7 derived meteorological data were used for all the v5.1 simulations presented here. As stated previously, different versions of WRF were used for the v5.0.2 and v5.1 simulations due to the updates made in both WRF and CMAQ (Section 2.1) that would have made performing the CMAQ simulations with output from the same version of WRF difficult and introduce some inconsistencies. While there were other updates made to WRF between versions 3.4 and 3.7, those changes were minor and did not impact the WRF results significantly for the configuration of the model used here.

Both WRF simulations employed the same options, which include the Rapid Radiation Transfer Model Global (RRTMG) long and short wave radiation (Iacono et al., 2008), Morrison microphysics (Morrison et al., 2005), and the Kain-Fritsch 2 cumulus parametrization (Kain, 2004). For the LSM and PBL models, the PX-LSM and ACM2 were used. Four-dimensional data assimilation (FDDA) was also employed in the WRF simulations. The namelists used for each WRF simulation are provided in the supplemental material (see S.4 and S.5). Model ready meteorological input files were created using version 4.1.3 of the Meteorology-Chemistry Interface Processor (MCIP; Otte and Pleim, 2010) for the WRF v3.4 data and MCIP version 4.2 (<https://www.cmascenter.org/help/documentation.cfm?model=mcip&version=4.2>) for the WRF v3.7 data.

Emission input data for the v5.0.2 simulation were based on version 1 (v1) of the 2011 modeling platform developed by the USEPA from regulatory applications (https://www.epa.gov/sites/production/files/2015-08/documents/lite_finalversion_ver10.pdf), while the base v5.1 simulation utilized emission data based on version 2 (v2) of the 2011 modeling platform. The v2 emission data became available after the completion of the v5.0.2 simulation using v1 emission data and it was determined that the latest version of the emission data would be used for the v5.1 simulation in order to obtain the best results. However, based on sensitivity simulations performed for January and July 2011 where the only difference was the emissions platform used, the most significant changes in the emissions between v1 and v2 (based on https://www.epa.gov/sites/production/files/2015-10/documents/nei2011v2_tsd_14aug2015.pdf) are highlighted below.

For the oil and gas sector, there were 4 major changes: 1) better aligning the inputs and emission factors between the EPA's Office of Atmospheric Program (OAP) work on the Greenhouse Gas (GHG) Emissions Inventory (EI) / GHG Reporting Program and the NEI on condensate tanks, liquids unloading, pneumatic devices and well completions, 2) additional information from the Western Regional Air Partnership (WRAP) based on new survey data and studies, 3) improved resolution of data (to county level rather than basin), and 4) new SCCs, including the distinction between Coal Bed Methane (CBM) wells from other natural gas (NG) wells. For other nonpoint sources, many states resubmitted data based on EPA or their own review of v1 (i.e. CA, CT, DC, DE, IA, ME, MI, MN, NC, NE, NY, OK, UT, VA, WA). Some tribes also submitted their data for the first time for the 2011 v2. For mobile sources, MOVES2014 was used in v2, while MOVES2010b was used for v1 (<https://www3.epa.gov/otaq/models/moves/moves-docum.htm>). Some commercial marine inventories were also updated. The differences in O_3 and $PM_{2.5}$ between those two simulations used were generally small and isolated, suggesting there is minimal impact to the comparison between the v5.0.2 and v5.1 simulations from the change in the emissions platform used. Figure S1 shows the impact on winter (January) and summer (July) O_3 and $PM_{2.5}$ between simulations using the different emission platforms.

With respect to fires in the NEI, wild land and prescribed fire emissions were altered for NC and DE. NC submitted their own emissions in going from v1 to v2, resulting in an over 95% reduction in NC wildfire emissions. Nationally, this caused emissions to be about 30% lower in 2011 v2 vs 2011 v1. The Delaware fire emissions were reduced about 96%, however the effects nationwide were small. For agricultural fires, updates from v1 resulted in a reduction of between 95-99% of emissions for WI, MI, OH, MO, and IL. Cumulatively, these changes reduced emissions about 34% nationwide.

The raw emissions files were processed using versions 3.5 (v1 emissions) and 3.6.5 (v2 emissions) of the Sparse Matrix Operator Kernel Emissions (SMOKE; <https://www.cmascenter.org/smoke/>) to create gridded, speciated hourly model-ready input emission fields for input to CMAQ. Electric generating unit (EGU) emissions were obtained using data from EGUs equipped with Continuous Emission Monitoring System (CEMS). Plume rise for point and fire sources were calculated in-line for all simulations (Foley et al., 2010; https://www.cmascenter.org/cmaq/documentation/4.7.1/INLINE_EMISSIONS_DEPV_NOTES.txt). Biogenic emissions were generated in-line in CMAQ using BEIS versions 3.14 for v5.0.2 and 3.61 (Bash et al., 2016) for v5.1. All the simulations employed the bi-directional ammonia flux (bi-di) option for estimating the air-surface exchange of ammonia, as well as the in-line estimation of NO_x emissions from lightning strikes.

Output from the various CMAQ simulations is paired in space and time with observed data using the Atmospheric Model Evaluation Tool (AMET; Appel et al., 2011). There are several regional and national networks that provide routine observations of gas and particle species in the U.S. The national networks include the EPA's Air Quality System (AQS; 2086 sites; <https://www.epa.gov/aqs>) for hourly and daily gas and aerosol PM species; the Interagency Monitoring of PROtected Visual Environments (IMPROVE; 157 sites; <http://vista.cira.colostate.edu/improve/>) and Chemical Speciation Network (CSN; 171 sites; <https://www3.epa.gov/ttnamti1/speciepg.html>) for daily average (measurements typically made every third or sixth day) total and speciated aerosol PM species; and the Clean Air Status and Trends NETwork (CASTNET; 82 sites; <http://www.epa.gov/castnet/>) for hourly O_3 and weekly aerosol PM species. In addition to these routinely available observations, the DISCOVER-AQ campaign (https://www.nasa.gov/mission_pages/discover-aq/) during July 2011 provides additional ground-based gas and aerosol PM measurements, along with unique aloft measurements made by aircraft, vertical profilers (e.g. Light Detection And Ranging (LiDAR) measurements), ozonesondes and tethered balloons (not utilized in this analysis however).

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4 Evaluation of major scientific improvements

In this section we evaluate the impact that several of the major scientific improvements in v5.1 have on the operational model performance. Unlike Foley et al. (2010), in which several individual major scientific improvements in CMAQ v4.7 were evaluated incrementally (e.g. each subsequent improvement is evaluated against the previous improvement), here we examine each scientific improvement separately by comparing simulations with the specific improvement removed (i.e. as it was in v5.0.2) to the base v5.1 simulation (CMAQv5.1_Base) which includes all the updates. While this has the disadvantage of not showing the incremental change in model performance due to each improvement, it does limit the number of simulations that need to be performed. In addition, it allows for easier examination of the effect of nonlinear increments on total model performance, as some updates to the modeling system may be affected by updates to other parts of the model, the effects of which on model performance may not be captured in an incremental testing format. Note that while some attempt is made to broadly identify the processes involved that cause the observed changes in model performance between v5.0.2 and v5.1, it would be too laborious (both to the reader and to the investigators) to comprehensively describe and investigate in-depth the processes involved that result in each observed difference in model performance described in this section. Where appropriate, the analyses presented in this section use the v5.0.2 base simulation (CMAQv5.0.2_Base) for comparison to the scientific improvement while for other improvements the v5.1 base simulation is used for comparison. In each case, the simulations being compared are noted. Table 2 provides a description of the CMAQ model simulations referred to in the following sections.

4.1 WRF and CMAQ meteorological updates

As discussed in section 2.1, there were several significant corrections/improvements made to the meteorological calculations in both WRF and CMAQ. While the focus of this work is on updates to the CMAQ model, certain options within WRF and CMAQ are linked, and therefore it is necessary to discuss the WRF model updates alongside the corresponding CMAQ model updates.

Figure 12 shows the cumulative impact that all the meteorological changes in WRF and CMAQ (i.e. changes to ACM2 and MOL) had on O_3 and $PM_{2.5}$ in January and July by comparing the CMAQv5.0.2_Base simulation to a CMAQv5.0.2 simulation using WRFv3.7 (CMAQv5.0.2_WRFv3.7) which includes the ACM2 and MOL updates. The effect of the changes on O_3 in January is mixed, with some areas (e.g. Florida, Chicago and the Northwest) showing a relatively large (2.5 ppbvppbV) increase in O_3 , while other areas (e.g. Southwest and Texas panhandle) show a relatively large decrease (-2.5 ppbvppbV) in O_3 . For $PM_{2.5}$, the differences in January are generally small and isolated, however there is a relatively large increase in $PM_{2.5}$ ($>2.5 \mu g m^{-3}$) in the San Joaquin Valley (SJV) of California due to the updates, which combined with the decrease in O_3 there as well, indicates a likely reduction in PBL height and mixing as the cause. There are also some relatively large decreases ($1.5 - 2.0 \mu g m^{-3}$) in $PM_{2.5}$ in the Northeast and around in the Great Lakes region (i.e. Chicago). Otherwise, most of the remaining impacts on $PM_{2.5}$ are relatively small ($< 1.0 \mu g m^{-3}$).

For July, the meteorological updates in WRF and CMAQ result in exclusively increased O_3 mixing ratios over land, which are considerably larger than the impacts observed in January. The largest increases ($4.0 - 10.0$ ppbvppbV) occur in the eastern U.S., particularly in the Southeast. Smaller increases of $2.0 - 4.0$ ppbvppbV occur across much of the U.S., while in the Gulf of Mexico and the Caribbean O_3 mixing ratios decrease roughly $2.0 - 6.0$ ppbvppbV across a large area. The difference in $PM_{2.5}$ in July is similar to that in January, with mostly small, isolated increases or decreases occurring in the eastern U.S. The largest increase ($2.0 - 2.5 \mu g m^{-3}$) occurs in the southern Ohio Valley (Kentucky and West Virginia), while the largest decreases ($> 2.5 \mu g m^{-3}$) occur in Louisiana and Texas (i.e. Houston).

It makes intuitive sense to see summertime O₃ mixing ratios increasing due to the meteorological changes in WRF and CMAQ, since the net effect of those changes was to increase mixing, particularly in the late afternoon and early evening, which in turn decreases the amount of NO titration of O₃ that occurs in the model, and ultimately results in higher O₃ mixing ratios on average.

Conversely, PM_{2.5} concentrations would be expected to decrease due to the increased mixing in the model, which would effectively decrease the concentrations of primary emitted pollutants (e.g. EC and OC), which was ~~generally seen~~observed in areas with the largest emissions (i.e. urban areas). In addition, changes in the oxidant (i.e. OH) concentrations would also potentially affect PM_{2.5} concentrations through increased or decreased SOA formation (spatial heterogeneity of PM_{2.5} formation)~~However, the spatial heterogeneity of PM_{2.5} formation in the atmosphere results which results in both~~spatially varying increases and decreases in PM_{2.5} concentrations.

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4.2 Aerosol updates

Several new SOA species from anthropogenic VOCs (i.e. AALK1, AALK2, APAH1, APAH2 and APAH3; Table 1) were added to AERO6 in v5.1 that ~~were~~are not present in v5.0.2. Figure 3 shows the difference in the monthly average sum total concentration of these five species for January and July 2011 between the CMAQv5.0.2_Base and CMAQv5.1_Base simulations. Since none of these species were present in v5.0.2, the difference totals in Figure 23 represent the additional SOA mass that these five species contribute to the total PM_{2.5} mass in v5.1. For both January and July, the monthly average concentration of these species is small, ranging between 0.0-0.1 µgm⁻³, with the largest concentrations in the eastern half of the U.S., particularly in the upper Midwest. However, the concentration of these new species during shorter time periods and smaller, isolated regions would be larger. In addition, the inclusion of these new species is potentially important for health related studies on the impact of PAHs. Since these species are not routinely observed and unique tracers have not been identified for alkene SOA, no comparison with observations is made here. Overall however, these new species represent a small addition to the total PM_{2.5} ~~concentration~~concentration in the model.

Along with the introduction of the new SOA species above, the pathways for the formation of acid enhanced isoprene SOA were also updated. The bottom panels in Figure 23 show the monthly average difference in the sum of the species containing isoprene SOA (AISO1, AISO2, AISO3 and AOLGB) between v5.1 and v5.0.2 (v5.1 – v5.0.2). For January, the difference in the sum of these species is relatively small, with minimum and maximum values peaking around ±0.5 µgm⁻³ consistent with the fact that isoprene emissions are low in winter. For July the difference is always positive (v5.1 higher than v5.0.2) and much larger compared to January, with peak differences exceeding 2.5 µgm⁻³, primarily in the areas with the highest aerosol SO₄²⁻ concentrations (i.e. Ohio Valley). Therefore, the updated IEPOX-SOA formation pathways in v5.1 represent a potentially significant contribution to the total PM_{2.5}, particularly during the summer. Increased isoprene emissions in v5.1 with BEIS v3.61 compared to v5.0.2 with BEIS v3.14 also contribute to the larger contribution of isoprene SOA in v5.1.

4.3 Cloud model and in-line photolysis updates

Changes in the photolysis/cloud model treatment in v5.1 have potentially significant impacts on the O₃ and PM_{2.5} estimates from the model. Figure 3 shows the difference in O₃ and PM_{2.5} for the CMAQv5.1_Base simulation and the CMAQv5.1_RetroPhot simulation (see Table 2 for simulation description). The CMAQv5.1_RetroPhot simulation is the same as the CMAQv5.1_Base simulation except it employs the same (old) photolysis/cloud model treatment as in v5.0.2. For January, O₃ mixing ratios (Figure

3a) and $PM_{2.5}$ concentrations (Figure 3c) are both higher across the Southeast and portions of California in the v5.1 simulation, indicating that v5.1 has much less photolysis attenuation due to the updates in the representation of cloud effects on photolysis.

The impact of the updated photolysis in v5.1 is considerably larger in July (when there is more convection) than in January. Peak O_3 differences in January were around 2.0 ppbv, whereas in July peak differences of greater than 5.0 ppbv (Figure 3b) occur over the Great Lakes (where low PBL heights can enhance the impact of changes in O_3). However, in general the difference in O_3 mixing ratios is larger in both magnitude and spatial coverage in July compared to January, indicating that the updated photolysis/cloud model treatment in v5.1 increases O_3 to a greater extent in July compared to January, as expected due to increased photolysis rates in the summer compared to winter. Overall, differences in O_3 in July range on average from 1.0 to 3.0 ppbv, with larger differences occurring in the major urban areas (e.g. Atlanta, Charlotte and Los Angeles) and off the coast of the Northeast corridor. The change in $PM_{2.5}$ is also larger (both in magnitude and spatial coverage) in July than January (Figure 3d). The greatest change is primarily confined to the eastern U.S., resulting in a roughly 0.1 to 0.5 $\mu g m^{-3}$ increase in $PM_{2.5}$ in v5.1, with the maximum increase located over the Great Lakes region and areas to the south, the result of increased SOA and gas-phase production of SO_4^{2-} due to greater OH^\cdot concentrations in v5.1.

Changes in the photolysis/cloud model treatment in v5.1 have potentially significant impacts on the O_3 and $PM_{2.5}$ estimates from the model. Figure 4 shows the difference in O_3 and $PM_{2.5}$ for the CMAQv5.1_Base simulation and the CMAQv5.1_RetroPhot simulation (see Table 2 for simulation description). The CMAQv5.1_RetroPhot simulation is the same as the CMAQv5.1_Base simulation except it employs the same (old) photolysis/cloud model treatment as in v5.0.2. For January, O_3 mixing ratios (Figure 4a) and $PM_{2.5}$ concentrations (Figure 4c) are both higher across the Southeast and portions of California in the v5.1 simulation, indicating that v5.1 has much less photolysis attenuation due to the updates in the representation of cloud effects on photolysis.

The impact of the updated photolysis in v5.1 is considerably larger in July (when there is more convection) than in January. Peak O_3 differences in January were around 2.0 ppbv, whereas in July peak differences of greater than 5.0 ppbv occur over the Great Lakes (where low PBL heights can enhance the impact of changes in O_3). However, in general the difference in O_3 mixing ratios is larger in both magnitude and spatial coverage in July compared to January, indicating that the updated photolysis/cloud model treatment in v5.1 increases O_3 to a greater extent in July compared to January, as expected due to increased photolysis rates in the summer compared to winter. Overall, differences in O_3 in July range on average from 1.0 to 3.0 ppbv, with larger differences occurring in the major urban areas (e.g. Atlanta, Charlotte and Los Angeles) and off the coast of the Northeast corridor. The change in $PM_{2.5}$ is also larger (both in magnitude and spatial coverage) in July than January, and is primarily confined to the eastern U.S. and results in a roughly 0.1 to 0.5 $\mu g m^{-3}$ increase in $PM_{2.5}$ in v5.1, with the maximum increase located over the Great Lakes region and areas to the south, the result of increased SOA and gas-phase production of SO_4^{2-} due to greater OH^\cdot concentrations in v5.1.

Additional diagnostic evaluation of photolysis/cloud model treatment in CMAQ was conducted based on the model predicted cloud albedo at the top of the atmosphere. The predicted cloud albedo from WRFv3.7, CMAQv5.0.2 and CMAQv5.1 were evaluated against cloud albedo from NASA's Geostationary Operational Environmental Satellite Imager product (GOES; <http://satdas.nsstc.nasa.gov/>). This evaluation was used to qualitatively determine if one CMAQ version better considers how clouds affect calculated photolysis rates. The GOES product has a 4km horizontal resolution and was re-gridded to the 12-km grid structure used in the WRF and CMAQ simulations using the Spatial Allocator utility (<https://www.cmascenter.org/sa-tools/>). The satellite data are available at 15 minutes prior to the top of the hour during daytime hours and were matched to model output at the top of the hour (see section S.1 in the supplemental material for further information). Figure 4 shows the average cloud albedo or reflectivity at the top of the atmosphere during daytime hours in July 2011 derived from the GOES

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satellite product (Figure 4a), and the cloud parameterizations within: WRF3.7 (Figure 4b), CMAQv5.1_RetroPhot (Figure 4c) and CMAQv5.1_Base (Figure 4d). Comparison of Figure 4b to 4c shows the dramatic differences between the clouds predicted by WRFv3.7 and the predictions from the cloud parameterization in v5.0.2. Most of these large differences, particularly over land, are now gone in model predictions from the CMAQv5.1_Base simulation which uses resolved clouds from WRF and sub-grid clouds from the convective cloud model within CMAQ (compare Figure 4b to 4d).

Two notable issues remain with the v5.1 modeled cloud parametrization. The photolysis cloud parameterization in v5.1 produces more clouds over water compared to the WRF parameterization, which is itself biased high for some parts of the Atlantic Ocean compared to GOES. This issue will be addressed by science updates planned for the CMAQ system and evaluation results are expected to improve in the next CMAQ release. A more significant issue, from an air quality perspective, is the under-prediction of clouds over much of the Eastern and West Central U.S. in the WRF predicted clouds, which is now directly passed along to CMAQ. This misclassification of modeled clear sky conditions can contribute to an over prediction of O₃ in these regions. Resolving this issue will require changes to the WRF cloud parameterization. Future research will also include changing the sub-grid cloud treatment currently used in the CMAQ system to be consistent with the sub-grid parameterization used in WRF. Section S.1 in the supplemental material provides a table with additional evaluation metrics of the modeled clouds over oceans versus over land and also describes how cloud albedo was calculated for the three model simulations.

~~Two notable issues remain with the v5.1 modeled cloud parametrization. The photolysis cloud parameterization in v5.1 produces more clouds over water compared to the WRF parameterization, which is itself biased high for some parts of the Atlantic Ocean compared to GOES. This issue will be addressed by science updates planned for the CMAQ system and evaluation results are expected to improve in the next CMAQ release (See Section 7.4). A more significant issue, from an air quality perspective, is the under-prediction of clouds over much of the Eastern and West Central US in the WRF predicted clouds, which is now directly passed along to CMAQ. This misclassification of modeled clear sky conditions can contribute to an over prediction of O₃ in these regions. Resolving this issue will require changes to the WRF cloud parameterization. Future research will also include changing the sub-grid cloud treatment currently used in the CMAQ system to be consistent with the sub-grid parameterization used in WRF. Section S.1 in the supplemental material provides a table with additional evaluation metrics of the modeled clouds over oceans versus over land and also describes how cloud albedo was calculated for the three model simulations.~~

4.4 Atmospheric chemistry updates

As detailed in section 2.4, numerous updates were implemented in the representation of atmospheric chemistry in v5.1. It would be extremely cumbersome to attempt to isolate the impact of each chemistry update individually. Instead, in order to assess the overall impact that the combined chemistry changes have on the model results, model comparisons are conducted using the CMAQv5.1_Base simulation, which employs the CB05e51 chemical mechanism (the v5.1 default chemical mechanism) and the CMAQv5.1_TUCL simulation (see Table 2 for description). The CMAQv5.1_TUCL simulation is the same as the CMAQv5.1_Base simulation except that it employs the CB05TUCL~~ue1~~ chemical mechanism (Whitten et al., 2010; Sarwar et al., 2012), the default mechanism in v5.0.2. Note that the aerosol updates discussed in section 4.2 were incorporated into the CB05e51 chemical mechanism (in the past that portion of the aerosol chemistry was separate from the gas-phase chemical mechanism). As

such, differences between the CMAQv5.1_TUCL and CMAQv5.1_Base simulations include impacts from those changes (i.e. Figure 2). In order to isolate primarily just the effect on PM_{2.5} from the atmospheric chemistry changes, the organic matter (AOMI; See S.2 and S.3 for species definition descriptions) mass has been removed from the comparisons of total PM_{2.5} mass discussed below.

Figure 5 shows the difference in monthly average O₃ and PM_{2.5} for January and July between the CMAQv5.1_Base and CMAQv5.1_TUCL simulations. For January, O₃ mixing ratios are higher in the simulation using the CB05e51 mechanism (CMAQv5.1_Base simulation), versus the CMAQv5.1_TUCL simulation, indicating generally higher O₃ during winter due to the updates in the CB05e51 mechanism. However, the overall impact of CB05e51 on O₃ is generally small (~2-4%), with maximum differences of only around approximately 1.0 ppbvppbv (~6%), primarily along the southern coastal areas of the U.S. PM_{2.5} is also higher in January in the simulation using the CB05e51 mechanism (CMAQv5.1_Base simulation), indicating higher PM_{2.5} with the CB05e51 mechanism versus CB05tuel, with the largest changes in PM_{2.5} primarily of 0.2 – 0.4 µgm⁻³ (~2-6%) primarily occurring in the eastern U.S., with differences ranging between 0.0 to – 0.4 µgm⁻³, with a notable larger isolated difference and greater than 1.0 µgm⁻³ (~6-8%) in the SJV of California (> 1.0 µgm⁻³) in the SJV.

For July, O₃ mixing ratios are higher across most areas in the CMAQv5.1_Base simulation (due to the CB05e51 mechanism), primarily across northern portions of the U.S., the Great Lakes region and in California (i.e. Los Angeles and the SJV). Most increases in O₃ in the CMAQv5.1_Base simulation range between 0.6 and 1.2 ppbvppbv (~2-4%), however larger increases of over 3.0 ppbvppbv (~4-8%) occur in southern California and over Lake Michigan (likely influenced in-part by low PBL heights over the lake). A small area of lower O₃ mixing ratios occurs off the eastern coast of the U.S. For PM_{2.5} in July, the difference in PM_{2.5} due to the CB05e51 chemical mechanism is relatively small, with differences in concentrations generally ranging from ±0.50 µgm⁻³ (~2-4%) across in the eastern U.S.

5 Evaluation of CMAQv5.1

In this section, comparisons are made of the operational performance of the CMAQv5.0.2_Base and CMAQv5.1_Base simulations by initially comparing the simulations to each other (model to model) and then evaluating them against a wide variety of available air quality measurements (see section 3). Several common measurements of statistical performance are used, namely mean bias (MB), mean error (ME), root mean square error (RMSE) and Pearson correlation. Note that representativeness (incommensurability) issues are present whenever gridded values from a deterministic model such as CMAQ are compared to observed data at a particular point in time and space, as deterministic models calculate the average outcome over a grid for a certain set of given conditions, while the stochastic component (e.g. sub-grid variations) embedded within the observations cannot be accounted for in the model (Swall and Foley, 2009). These issues are somewhat mitigated for networks that observe for longer durations, for example the CSN and IMPROVE networks which are daily averages and the CASTNET observations which are weekly averages. The longer temporal averaging helps reduce the impact of stochastic processes, which can have a large impact on shorter (e.g. hourly) periods of observation (Appel et al., 2008).

There are several important differences to keep in mind between the comparison of the CMAQv5.0.2_Base and CMAQv5.1_Base simulations beyond the obvious changes to the model process representations discussed in the previous sections. First, the simulations use different versions of WRF (as discussed in Sections 2.2 and 4.1). This was intentional, as it was determined that

the changes made from WRF v3.4 (used in the CMAQv5.0.2_Base simulation) to WRF v3.7 (used in the CMAQv5.1_Base simulation) and subsequent required changes made to the CMAQ code represent a change to the overall WRF-CMAQ modeling system and therefore should be evaluated together. Second, the emission inventories for the two base runs are slightly different, as discussed in Section 3. While the changes between the emission inventories are generally minor, they do represent another difference between the simulations, and where possible the effect of the different inventories on the model performance is noted. Finally, it should also be noted that the windblown dust treatment was employed in the CMAQv5.0.2_Base simulation but not in the CMAQv5.1_Base simulation. This was due to issues with the implementation of the updated windblown dust treatment in v5.1 that were not discovered until after the model was released and the CMAQv5.0.2_Base simulation was completed. However, the contribution of windblown dust to total PM_{2.5} in v5.0.2 tends to be small and episodic and therefore should not constitute a significant impact to the performance differences between v5.0.2 and v5.1, especially for the monthly averages generally shown here. For reference, the v5.0.2 simulated seasonal average values of PM_{2.5} and maximum daily 8-hr average (MDA8) O₃ are provided in Figures S2 and S3 respectively.

5.1 PM_{2.5}

Figure 6 shows the seasonal average difference in model simulated PM_{2.5} between v5.0.2 and v5.1 (CMAQv5.1_Base – CMAQv5.0.2_Base), with cool colors indicating a decrease in PM_{2.5} in v5.1 (versus v5.0.2) and warm colors indicating an increase in PM_{2.5}. Figure 7 shows the seasonal mean bias (MB) for PM_{2.5} for the CMAQv5.1_Base simulation, while Figure 8 shows the change in the absolute value of the seasonal mean bias (|MB|) in PM_{2.5} between the CMAQv5.0.2_Base and CMAQv5.1_Base simulations. Cool colors indicate smaller PM_{2.5} |MB| in the CMAQv5.1_Base simulation (versus the CMAQv5.0.2_Base simulation), while warm colors indicate larger |MB| in the CMAQv5.1_Base simulation.

During winter, v5.1 predicts lower PM_{2.5} concentrations in the eastern U.S. and portions of western Canada compared to v5.0.2, but higher PM_{2.5} concentrations in the SJV and isolated portions of Mexico and Alabama (due to emissions changes affecting that state) (Figure 6). Almost all the differences in PM_{2.5} are due to changes in the emissions between the two simulations (see Figure S1a). PM_{2.5} is largely overestimated in the eastern U. S. and underestimated in the western U.S. in the winter in CMAQv5.1_Base simulation (Figure 7a). The change in |MB| between v5.0.2 and v5.1 is negative (reduced MB in v5.1) across the majority of the sites, with relatively large reductions (3–5 μgm⁻³) in |MB| in the Northeast, upper Midwest (i.e. Great Lakes region), SJV, and portions of the mid-Atlantic (e.g. North Carolina) (Figure 8a). Alabama is a notable exception, with the MB increasing in the v5.1 simulation due to changes in the emissions inventory. Figure S1S4 presents a histogram of the change in PM_{2.5} |MB| using the same data and color scale as in Figure 8. It's clear from the histogram the large percentage (69.2%) of sites where the |MB| decreases in the v5.1 simulation in the winter (Figure S1S4a), demonstrating a significant improvement in the PM_{2.5} performance for v5.1 versus v5.0.2.

The diurnal profile of PM_{2.5} for winter (Figure S29a) shows a relatively large decrease in MB throughout most of the day in the v5.1 versus v5.0.2, particularly during the overnight, morning and late afternoon hours. A similar improvement is seen in the RMSE, while the correlation also improves for all hours (Figure S5). Finally, Figure 910 shows seasonal and regional stacked bar plots of PM_{2.5} composition (SO₄²⁻, NO₃⁻, NH₄⁺, EC, OC, sSoil, NaCl, NCOM, and PM-Other). sWhere Soil is based on the IMPROVE soil equation and contains both primary and secondary sources of soil (Appel et al., 2013), and, while PM-Other represents the unspicified PM mass in the inventory (see Appel et al., 2008) The five regions shown in Figure 109 are Northeast (MaineE, New HampshireE, VermontE, MassachusettsA, New York, New Jersey, MDaryland, DelawareE, ConnecticutE, Rhode

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Island, Pennsylvania, District of Columbia, Virginia and West Virginia), Great Lakes (Ohio, Michigan, Indiana, Illinois and Wisconsin), Atlantic (North Carolina, South Carolina, Georgia and Florida), South (Kentucky, Tennessee, Mississippi, Alabama, Louisiana, Missouri, Oklahoma and Arkansas) and West (California, Oregon, Washington, Arizona, Nevada, New Mexico). These regions are derived from principle component analysis to group states with similar PM_{2.5} source regions together. For winter, the total PM_{2.5} high bias is reduced across all five regions, with most of the improvement coming from primary emitted species such as EC and OC, non-carbon organic matter (NCOM; Table S.2 or S.3) and PM-Other, indicating that improvements in the representation of mixing under stable conditions helped in reducing the high bias. Still, a large bias remains for OC, which may be due to an overestimation of the residential wood combustion in the NEI.

For spring, the changes in PM_{2.5} are much more isolated than in winter (Figure 6b), with the largest decreases occurring around Montreal (Canada), North Carolina, and portions of the desert Southwest (lack of wind-blown dust in v5.1 likely contributes to this decrease in the desert Southwest). A small increase in PM_{2.5} is again noted in Alabama due to emissions changes between the v1 and v2 emissions. The MB for PM_{2.5} in the spring is relatively small, with most sites showing having an underestimation of 1-3 µg m⁻³, while some overestimations of PM_{2.5} occur in the Northeast, Great Lakes and Northwest coast (Figure 7b). As expected with the relatively small changes in PM_{2.5} concentrations in spring, the difference in |MB| does not change significantly between v5.0.2 and v5.1 is relatively small, with most changes differences in |MB| less than ±1.0 µg m⁻³ (Figure 8b). Some slightly larger decreases in |MB| occur in the Northeast and Alabama, while some larger increases in |MB| occur in the desert Southwest. About half (49.8%) of the sites show an improvement in |MB| (Figure S4S3b). The diurnal profile of PM_{2.5} for spring (Figure S3) shows a consistent underestimation of PM_{2.5} throughout most of the day in the v5.0.2 simulation, which becomes larger in the v5.1 simulation with the overall decrease in PM_{2.5} in the spring (Figure 9b). However, the RMSE is lower during the overnight, morning and afternoon hours in the v5.1 simulation, while the correlation also improves throughout most of the day, the exception being 1pm to 4pm LST (Figure S6). Total PM_{2.5} MB improves in three of the five regions shown in Figure 9, with most of the improvement again coming from reductions in the primary emitted species.

In the summer, PM_{2.5} is considerably higher (> 5.0 µg m⁻³) across the eastern U.S. in the CMAQv5.1_Base simulation, particularly in Mississippi, Alabama, Georgia and portions of the Ohio Valley, while PM_{2.5} is lower in isolated areas in eastern North Carolina, Montreal, Canada and small areas in the southwest U.S. and Mexico (Figure 6c). The changes in PM_{2.5} in Alabama, Montreal and eastern North Carolina are all related, at least in part, to changes in the emissions between v1 and v2 (see Figure S1b). The increase in PM_{2.5} is primarily due to the updates to the IEPOX-SOA chemistry in v5.1 (Figure 2), updates to BVOC emissions in BEIS v3.61 (approximately 1.0 µg m⁻³ increase PM_{2.5} in the southwest U.S.), and the ACM2/MOL updates in WRF and CMAQ (Figure 1), with smaller contributions from the updates in CB05e51 chemical mechanism (Figure 5) and updates to the clouds/photolysis (Figure 3). Despite the increase in PM_{2.5} with v5.1, PM_{2.5} remains largely underestimated in the summer, with largest underestimations in the southeast U.S. and California (Figure 7c). However, the result of the widespread increase in PM_{2.5} in the CMAQv5.1_Base simulation is a similar large, widespread reduction in the |MB| across the eastern U.S., particularly in the Southeast (except eastern North Carolina and Florida) and the Ohio Valley, where reductions in |MB| range from 3.0 - 5.0 µg m⁻³ (Figure 8c). Smaller increases in the |MB| (typically less than 2.0 µg m⁻³) occur in eastern North Carolina and Florida, and isolated areas in the western U.S. Of all the sites, 60.4% showed an improvement in |MB|, with a large number of sites showing reductions in |MB| greater than 5.0 µg m⁻³ (Figure S4S3c). PM_{2.5} is underestimated throughout the day in both the v5.0.2 and v5.1 simulations (Figure 9cS4) in summer, with the underestimation improving slightly in v5.1, particularly during the afternoon and overnight hours. RMSE improves during the daytime hours in v5.1, while correlation is significantly higher in v5.1 than v5.0.2

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throughout the entire day (Figure S7). Total PM_{2.5} is underestimated by the model in four of the five regions (West region being the exception), but improves in three of those four regions with v5.1, with small increases in SO₄²⁻ and NH₄⁺, and larger increases in OC and NCOM contributing to the improvement (Figure 109).

For the fall, the difference in PM_{2.5} between v5.0.2 and v5.1 is again small (very similar to the spring), with the largest increases occurring in Alabama and western Canada (both emissions related) and the largest decreases occurring in Montreal, Mexico and isolated areas in the eastern and Midwest U.S. (Figure 6). The overall pattern in MB is somewhat similar to that of the spring (Figure 7d), with relatively small MBs in the Eastern U.S. ($\pm 2.0 \mu\text{gm}^{-3}$) and larger MBs along the west coast (underestimated in California and overestimated in the Northwest). As expected, the change in the |MB| between v5.0.2 and v5.1 is also relatively small in the fall, with the majority of the sites having a change in |MB| of less than $\pm 2.0 \mu\text{gm}^{-3}$ (Figure 8d), while 65.3% of the sites show a reduction in |MB| (Figure S4S7d). The average diurnal profile of PM_{2.5} in the fall (Figure 9d) is similar to the spring, with improved MB in v5.1 during the overnight, morning and late afternoon/evening hours and lower RMSE and higher correlation throughout the entire day (Figure S5S8). Total PM_{2.5} is overestimated in all five regions in the fall (Figure 910), but improves in v5.1 in four of those regions (exception being the South), with decreases in the primary emitted species responsible for most of the improvement.

5.2 Ozone

For the winter, O₃ widely decreases in the CMAQv5.1_Base simulation versus the CMAQv5.0.2_Base simulation across the western U.S., with the seasonal average decreases ranging between 1.0 – 3.0 ppbvppbV, and several areas where decreases exceed 3.0 ppbvppbV, primarily over the oceans (Figure 11a9). In the eastern U.S., the change in O₃ is relatively small and isolated, the exception being along the coast of Louisiana (due to differences in emissions; see Figure S1c)A and a small portion of Florida, where increases in O₃ exceed 5.0 ppbvppbV. Ozone is underestimated at most sites across the northern portion of the U.S., with the largest underestimations occurring in Colorado, Wyoming and Utah. Despite the decreases in O₃ with v5.1, O₃ is still overestimated in Florida, along the Gulf Coast of Mexico, the Southwest U.S. and in California (Figure 112a). There is a widespread reduction in the O₃ |MB| in California and increased |MB| in the upper Midwest with v5.1, while across the rest of the domain the change in |MB| is relatively small and mixed in direction (Figure 123a). The majority of the change in O₃ falls between $\pm 5.0 \text{ ppbvppbV}$, with slightly more sites (55.8%) showing a reduction than increase in |MB| (Figure S96a). The average diurnal profile of O₃ in the winter (Figure S714a) shows slightly lower mixing ratios during most of the day, the exception being the late afternoon and early evening hours when mixing ratios are slightly higher. The result is lower-reduced MB and RMSE, and higher correlation throughout the day with v5.1 versus v5.0.2 (Figure S10). The NO_x diurnal profile also generally improves throughout the day in winter (Figure 15a), with decreases in MB and RMSE in the afternoon/early evening and increased correlation throughout the day (Figure S811).

The pattern of change in O₃ between v5.0.2 and v5.1 in spring is similar to winter, with lower O₃ mixing ratios in the western U.S. and higher mixing ratios in the eastern U.S. in v5.1 compared to v5.0.2 (Figure 101b). Decreases in O₃ mixing ratios in the western U.S. in v5.1 range from roughly 1.0 – 3.0 ppbvppbV (similar to winter), while in the eastern U.S. the increases generally range from 1.0 – 2.0 ppbvppbV, with isolated areas of larger increases. The MB of O₃ for the v5.1 simulation primarily ranges from slightly over- to slightly underestimated across most the sites, with larger overestimations along the Gulf Coast and larger underestimations in the western U.S. (Figure 112b). The change in |MB| between v5.0.2 and v5.1 shows mixed results (Figure

123b), with slight increases and decreases across much of the eastern U.S. and a relatively large increase in |MB| in the Midwest (Colorado and Wyoming). The |MB| mostly improved across the Gulf Coast and in California due to reduced O₃ mixing ratios from the new marine halogen chemistry and enhanced O₃ deposition to ocean surfaces. Roughly half (49.4%) of the sites showed a reduction in |MB| (Figure S6b) with v5.1. The diurnal profile of O₃ for spring (Figure 14bS9) shows a relatively large increase in mixing ratios improvement in MB in v5.1 in the late afternoon and evening (4pm to 10pm LST), resulting in a large improvement in MB during that time (Figure S12), with similar improvements are noted in RMSE and slightly higher and correlations in the afternoon and evening hours. The NO_x diurnal profile also shows a large decrease in the late afternoon and early evening mixing ratios (Figure 15b), with a large decrease in both MB and RMSE during that time, improved representation of the morning rush-hour peak, and improved correlation improves throughout the day as well (Figure S13S14).

For the summer, the pattern of change in O₃ is markedly different from the winter and spring, with widespread, large increases in O₃ mixing ratios across the eastern U.S. and decreases in O₃ mixing ratios in the Gulf of Mexico (partially due to differences in emissions; see Figure S1d), southern Florida and over the eastern Atlantic (Figure 110c) ocean. Increases in O₃ in v5.1 in the eastern U.S. range from 2.0 – 10.0 ppbvppbV, with isolated areas of larger increases in the major urban areas (e.g. Chicago, Illinois and Atlanta, Georgia) that can largely be attributed to the updates to the ACM2 and the MOL calculation in WRF and CMAQ (Figure 2b) as well as increased photolysis in v5.1 versus v5.0.2 (Figure 1). Smaller increases in O₃ occur in the western U.S., particularly southern California and the SJV. Decreases in O₃ over the oceans are also large as a result of the introduction-inclusion of a chemical sink for O₃ due to marine halogen chemistry in v5.1 (Sarwar et al., 2015), with some decreases exceeding 10.0 ppbvppbV. The MB of O₃ for the v5.1 simulation shows widespread overestimations in the eastern U.S., particularly along the Gulf of Mexico, while in the western U.S. the MB is mixed, with the largest overestimations occurring along the California coast (Figure 142c).

As expected, the consequence of the widespread increase in O₃ in the eastern U.S. in v5.1 is a corresponding widespread increase in the |MB| compared to v5.0.2, particularly in the Mid-Atlantic and Southeast (Figure 123c). Ozone |MB| decreases along the coast of Florida and along the Gulf of Mexico, likely the result of decreased O₃ over the water, from changes in the deposition of O₃ over water and the inclusion of the halogen chemistry update. The change in |MB| in the western U.S. is mixed, with some areas showing improved |MB| (e.g. SJV), while others show increased |MB| (e.g. southern California). And while the diurnal profiles of O₃ show that MB mixing ratios increases throughout much most of the day in v5.1 (exception being 12am – 5am) (Figure 14c), resulting in increased MB and RMSE throughout the morning and early afternoon hours (Figure S14). However, RMSE decreases substantially during the late afternoon and overnight hours, and the correlation improves throughout the entire day (Figure S14). The NO_x concentrations are lower throughout the day within v5.1 compared to v5.0.2 (Figure 15c), which results in large improvements in the MB in the morning and afternoon periods and slightly increased MB in the middle of the day, while RMSE and correlation improve throughout the day (Figure S125).

For the fall, the pattern of change in O₃ for v5.1 versus v5.0.2 is nearly identical to spring (Figure 11d0), with widespread decreases in O₃ in the western U.S. (possibly due to reduced cloud mixing and entrainment from the free troposphere) and mostly small increases in O₃ in the eastern U.S., with the exception of larger increases in several of the major urban areas (e.g. St. Louis, Missouri and Atlanta, Georgia). The changes are generally small, between ±2.0 ppbvppbV, with isolated areas of larger increases or decreases. Ozone is also lower over the Pacific and Atlantic oceans and the Gulf of Mexico. While the change in O₃ between v5.0.2 and v5.1 very similar to the spring, the MB pattern for v5.1 is not. Unlike the spring where O₃ was largely underestimated, in the

fall O₃ ~~continues to be~~ overestimated across most of the sites, much like the summer (Figure 12d). The Midwest sites ~~have~~ shows lowest overall MB, while the east and west coasts ~~have~~ show large overestimations of O₃. The increased O₃ in the eastern U.S. with v5.1 results in generally higher |MB| compared to v5.0.2, while in the western U.S. the result is slightly lower |MB| on average, the exception being southern California (Figure 12d). As was the case in the spring, slightly less than half the sites (48.9%) showed a reduction in |MB|, with the majority of the change falling between ± 5.0 ppbv (Figure S6d). The diurnal profile of O₃ in the fall shows increased ~~mixing ratios~~ MB in v5.1 ~~compared to v5.0.2 throughout most of the day during the daytime hours and slightly decreased mixing ratios overnight~~ (Figure 14d), ~~resulting in increased MB during the daytime and lower MB overnight. However, the~~ but again lower RMSE is lower and the correlation is higher throughout the day ~~and higher correlation throughout the entire day~~ (Figure S13f). Similar to the other seasons, the diurnal profile of NO_x in the fall shows lower ~~mixing ratios~~ throughout the day (Figure 15d), particularly in the early morning and late afternoon hours, ~~resulting in higher MB in the morning and lower MB in the afternoon, and lower~~ while RMSE is lower and higher correlation is higher throughout the entire day ~~in~~ with v5.1 (Figure S14f).

5.2.3 ANs, PN_s and Comparisons to Aircraft Measurements

Previous studies have shown that CMAQ can significantly overestimate NO_y mixing ratios (e.g. Anderson et al., 2014). To help address the NO_y overestimation in CMAQ, updates were made to the atmospheric chemistry in v5.1 pertaining to the formation and cycling of alkyl nitrates (ANs), peroxy nitrates (PNs) and NO_y in the model (Section 2.4.1). Overall, monthly average hourly NO_y mixing ratios at AQS sites decreased between approximately 13% (January) and 21% (July) in the CMAQv5.1 Base simulation versus the CMAQv5.0.2 Base simulation. The result is a slight decrease in the normalized mean error (NME) in January from 70% (v5.0.2) to 61% (v5.1), but a much larger decrease in NME in July from 151% (v5.0.2) to 101% (v5.1). Mixing ratios of ANs and PN_s are not routinely measured, however ~~In addition to the routine ground-based measurements, the DISCOVER-AQ campaign~~ (https://www.nasa.gov/mission_pages/discover-aq/) ~~campaign~~ that took place over the Baltimore, Maryland and Washington, D.C. area in July 2011 provides ~~aircraft~~ a unique measurements dataset containing both ground-based and upper air (i.e. aircraft) measurements. ~~Not only does this allow evaluation of the model performance throughout the PBL, the unique measurements also allow evaluation against species that are not routinely observed, specifically of peroxy nitrates (PNs) and alkyl nitrates (ANs), both of which are important species in along with NO₂, NO_y, HNO₃ and O₃ chemistry.~~ The National Oceanic and Atmospheric Administration (NOAA) P3B aircraft performed measurement flights on a number of days during the DISCOVER-AQ campaign. Those flights included vertical spirals over several locations, one of which was Edgewood, MD (39.41°N, 76.30°W; 11m above sea level), a site that often measures very high O₃, and in recent years has measured some of the highest O₃ in the eastern U.S.

Figure 13f shows vertical profiles of observed and CMAQ (v5.0.2 and v5.1) simulated O₃, NO₂, NO_y, ANs, PN_s and ~~nitric acid~~ (HNO₃) for the Edgewood site on July 5, 2011. ~~Several spirals were performed over the Edgewood site that day, roughly taking place in the late morning, early afternoon and late afternoon, so the profiles shown represent an average profile throughout the day.~~ While O₃ is underestimated throughout the PBL by both versions of the model on that day, the underestimation ~~is significantly~~ improves ~~significantly~~ in the v5.1 simulation. NO₂ and NO_y are overestimated throughout the PBL by both versions of the model, but again, the overestimation is greatly improved in the v5.1 simulation. The PN_s, ANs, and HNO₃ show mixed results, with the ANs performance improving, the PN_s performance degrading and the HNO₃ performance relatively unchanged with v5.1. Note that there has been an update in the recommended PAN formation and degradation equilibrium constant (<http://iupac.pole-ether.fr>) which lowers the predicted PAN concentrations in CMAQ and is currently being examined for its impact on other species. On this

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particular day, v5.1 generally shows a large improvement in performance over v5.0.2, however the results on other days may be different, but it does highlight the large change in NO_y mixing ratios that can be expected with v5.1.

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6 Modeled Response to Emission Changes

One of the primary applications of air quality models is to determine the impact that changes (e.g. reductions from abatement strategies) in emissions have on ambient air quality. Examples of this type of application include Federal rules and State Implementation Plans (SIPs) which aim to reduce emissions (through regulations) in order to meet mandated air quality standards. In this type of application, the air quality model is run using both baseline (often current year) and future year emissions (when emissions are typically lower due to state and national regulatory efforts) and then the change in criteria pollutant (e.g. O_3 and $\text{PM}_{2.5}$) concentrations between the two simulations is quantified in order to assess the impact (benefit) that emission reductions will have on future ambient air quality. As such, it is important to establish the ability of the model to accurately simulate the future ambient air quality given a known change in emissions, which here is referred to as the model responsiveness (to emission changes).

Some previous analyses comparing observed changes in ambient air quality (over periods witnessing large reductions in emissions) to CMAQ estimated changes in ambient air quality (with estimated reductions in emissions) during the same period have shown that the model tends to underestimate the observed change in ambient O_3 , suggesting the model may be under-responsive to the emission reductions impacting O_3 (Gilliland et al., 2008; Foley et al., 2015). The over/under responsiveness of the model to emission projections can have implications in the planning process for determining the extent to which emissions must be reduced in order to meet future air quality standards. In the following sections, we examine the model responsiveness to emission reductions in CMAQ v5.0.2 and v5.1 by computing the ratio of maximum daily 8-hr average (MDA8) O_3 mixing ratios and total $\text{PM}_{2.5}$ (and select $\text{PM}_{2.5}$ component species) between simulations using the base emissions inventories and those employing 50% reductions in NO_{x+o} -VOC and SO_x and SO_2 emissions in order to estimate a model responsiveness to the emission reductions for each version of the model. The model responsiveness for v5.1 is then compared to that of v5.0.2 to determine whether the model responsiveness increased, decreased or was unchanged in the new version of the model.

6.1 O_3

Figure 147 shows the difference in the ratio (emission cut simulation / base simulation) of MDA8 O_3 for the 50% cut in anthropogenic NO_{x+o} and VOC scenarios, binned by model MDA8 O_3 mixing ratio. Values greater than zero indicate v5.1 is more responsive to the NO_{x+o} or VOC cut than v5.0.2, while values less than zero indicate v5.1 is less responsive than v5.0.2. For both January and July, the median difference in ratio values for all bins for the 50% NO_{x+o} cut scenario are greater than zero, indicating that v5.1 is more responsive than v5.0.2 to the cut in NO_{x+o} . For the 50% cut in VOC emissions the difference in the ratio values is mixed across the two months and the different bins. For January, all of the bins indicate that v5.0.2 is more responsive than v5.1 to the 50% VOC cut, with the greatest difference occurring for MDA8 O_3 mixing ratios greater than 65 ppbvppbV. For July, v5.1 is slightly more responsive to the VOC cut for MDA8 O_3 mixing ratios less than 75 ppbvppbV and less responsive for MDA8 O_3 mixing ratios greater than 85 ppbvppbV.

6.2 $\text{PM}_{2.5}$

Figure 158 shows the difference in the -ratio (emission cut simulation / base simulation) of PM_{2.5} and select PM_{2.5} component species between v5.0.2 (blue) and v5.1 (red) for January (top) and July (bottom) for a 50% cut in anthropogenic emissions of NO_x, VOC and SO_x. For January, the overall response of modeled PM_{2.5} (PMI) to a 50% reduction in NO_x is primarily driven by a decrease in nitrate and its associated ammonium. CMAQ v5.1 PM_{2.5} is slightly less responsive to NO_x reductions compared to v5.0.2, but is still overall quite similar. The VOC cut shows greater response with v5.1 than v5.0.2 in January in ANCOMIJ (non-carbon organic matter attached to primary organic carbon; Simon and Bhawe, 2012), AUNSPECIJ (unspeciated PM), AOMIJ (all organic matter), AORGAJ (SOA from anthropogenic VOCs) and AORGBJ (SOA from biogenic VOCs) and total PM_{2.5} (see species definition files in supplemental material S.2 and S.3). Note that the letters I and J after the species name indicate which CMAQ modal distributions are being included in the total species mass, with I indicating the Aitken mode and J indicating the Accumulation mode. Since NCOMIJ is nonvolatile, its change reflects how reducing VOCs changes oxidants such as OH. In general, the model PM_{2.5} is not very sensitive to VOC cuts in January. And finally, for the 50% SO_x cut scenario, PM_{2.5} is only slightly less responsive with v5.1, with all the species being similarly responsive to the SO_x cut using v5.1 compared to v5.0.2.

For July, the NO_x cut scenario with v5.1 shows greater responsiveness for the ASO4IJ (sulfate), ANH4IJ (nitrate), AECIJ (elemental carbon), APOAIJ (primary organic aerosol), AORGCJ (SOA from glyoxal and methylglyoxal processing in clouds) species and total PM_{2.5} versus v5.0.2. For the VOC cut scenario, the AORGAJ species show increased responsiveness with v5.1. CMAQ-v5.1 alkane SOA is not dependent on NO_x levels or HO₂:NO ratios, so the decrease in VOC precursors have a more direct effect than for the aromatic systems (the only AORGAJ in v5.0.2), where decreasing the VOC precursors can also modify the HO₂:NO ratio and thus yields. CMAQ v5.1 PMIJ becomes slightly more responsive to SO_x cut as a result of an increased sensitivity of biogenic SOA to sulfur containing compounds. This link results from the IEPOX acid-catalyzed SOA in the model which has been shown to be correlated with sulfate (Pye et al., 2016 in-prep).

7 DiscussionSummary

A new version of the CMAQ model (v5.1) containing numerous scientific updates has been released and evaluated in terms of the change in operational performance against the previous version of the model (v5.0.2), performance compared to observations, e and response to changes in inputs (i.e. emissions). Specifically, updates were made to the ACM2 scheme in both WRF and CMAQ to improve the vertical mixing in both models, along with updates to the MOL calculation, which also directly impacted the vertical mixing in the WRF-CMAQ system. The overall net effect of these updates was to increase the ventilation in the model, particularly during the transition periods (morning and evening), which in turn reduced the concentration of primary emitted species (e.g. NO_x and OC) and consequently increased simulated O₃ (a result of reduced NO_x titration) and decreased PM_{2.5} concentrations due to greater dilution. Several new SOA formation pathways and species were added to v5.1, resulting in increased SOA, particularly in the southeast U.S., and improved PM_{2.5} performance in the summer, as PM_{2.5} is often-generally underestimated by CMAQ during the summer in the U.S.

The in-line photolysis model within CMAQ was updated in v5.1. Cloud cover for the photolysis model in v5.0.2 used a single cloud deck with a constant cloud fraction and water droplet mixing ratio. In v5.1, multiple cloud decks with variable cloud fractions and multiple types of water condensates are used in the photolysis model to be more consistent with the WRF meteorological model and the CMAQ cloud model. The net effect of this change was to decrease the amount of sub-grid clouds in the photolysis calculation in v5.1, which in turn results in higher photolysis rates and thus higher predicted O₃ mixing ratios on average. In

addition to the change to the photolysis model, the refractive indices for aerosol species are now both wavelength and composition dependent. Changes in aerosol scattering and extinction also introduce ~~run-time~~ options for how to calculate their optical properties and allow the user to specify which aerosol mixing model and method to use to solve the Mie scattering theory. The atmospheric chemistry in the model has also been updated from CB05~~TUCLfuel~~ to CB05e51 in v5.1, which includes, among other things, updates to the NO_x reactions, additional isoprene extensions, explicit representation of several HAPs, and a simple parameterization of the effects of halogens on O₃ in marine environments. The net effect of going from CB05~~TUCLfuel~~ to CB05e51 was to increase O₃ in the winter and summer, while increasing PM_{2.5} slightly in the winter and increasing/decreasing PM_{2.5} slightly in the summer.

Overall, the scientific updates in v5.1 resulted in relatively dramatic improvements in model performance for PM_{2.5} in the winter and summer and small overall changes in performance during spring and fall. PM_{2.5} concentrations decreased significantly in v5.1 in the winter when PM_{2.5} is typically overestimated by CMAQ over the U.S., and increased significantly in the summer when PM_{2.5} is typically underestimated by the model. The change in O₃ mixing ratios in v5.1 resulted in mixed improvement in MB, both spatially and temporally, with the summer showing the largest increase in MB. However, RMSE largely improved regardless of season and showed a larger improvement spatially across the sites than MB, and the correlation is almost always higher with v5.1. Comparisons of vertical profiles of several species taken over Edgewood, MD ~~on July 5, 2011~~ during the DISCOVER-AQ campaign showed improved performance within v5.1 throughout the PBL for O₃, NO₂, NO_y, ANs and CO, with the PNs being the only species to show degraded performance on that day. ~~And while the complexity of the model increased (e.g. additional species and reactions), the computational time required to complete a v5.1 simulation remained similar to v5.0.2 due to several improvements made to the model code to increase computational efficiency.~~

The response of the model to changes in emission inputs was examined by comparing the ratio of the base v5.0.2 and v5.1 simulations to sensitivity simulations with 50% cuts each to anthropogenic NO_x-VOC and ~~SO_x eSO_x~~ emissions. CMAQv5.1 simulated MDA8 O₃ exhibited more responsiveness (greater reduction) to the 50% NO_x cut in January and July than v5.0.2, which is considered an improvement as previous studies suggested CMAQ O₃ to be under-responsive to large changes in emissions. The responsiveness of PM_{2.5} to the emission cuts is more complicated than for O₃ since there are many more species comprising PM_{2.5} and some of those have greater or smaller response with v5.1. However, the new pathways of formation for several PM_{2.5} components in v5.1 generally result ~~+3+~~ in greater responsiveness in v5.1 compared to v5.0.2 for the various emission cut scenarios.

Finally, a number of important science updates are in development and will be available in the next release of CMAQ (v5.2), ~~which update or correct known issues in v5.1, and which~~ improve upon the existing science in the model. These updates include a new version of the windblown dust treatment (Foroutan et al., 2016), the Carbon-Bond 6 (CB6) chemical mechanism (Ramboll Environ, 2016), enhancements to the calculation of semi-volatile Primary Organic Aerosol (POA) and SOA from combustion sources in CMAQ (Pye et al., 2016), and additional updates to the calculation of clouds. In addition to the model updates, a number of instrumented versions of the model (e.g. decoupled direct method, sulfur tracking) will also be released with v5.2. These updates represent potentially significant improvements over the current options in v5.1 (specifically the updated windblown dust treatment) and therefore are being made available to the community more quickly than they might have in the past.

Code availability

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CMAQ model documentation and released versions of the source code, [including all model code used in this study](#), are available at www.cmaq-model.org. The updates described here, as well as model post-processing scripts, are available upon request. [The WRF model is available for download through the WRF website \(<http://www.wrf-model.org/index.php>\)](#).

Data availability

- 5 The raw observation data used are available from the sources identified in Section 3, while the post-processed observation data are available upon request. The CMAQ model data utilized are available upon request as well.

Disclaimer

The views expressed in this article are those of the author[s] and do not necessarily represent the views or policies of the U.S. Environmental Protection Agency.

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Table 1: New/revised SOA species in the CMAQ v5.1 AERO6 mechanism.

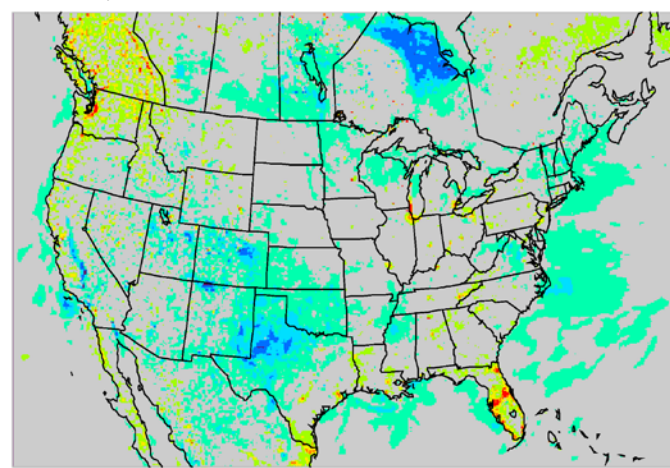
Aerosol Species	Change since v5.0.2	Applicable Mechanism	Description of Modification
AH3OP	added	all	Hydronium ion (predicted by ISORROPIA for I+J modes), used for IEPOX uptake
APAH1,2	added	cb05e51, saprc07tb, saprc07tc, saprc07tic, racm	Naphthalene aerosol from RO ₂ +NO reactions
APAH3	added	cb05e51, saprc07tb, saprc07tc, saprc07tic, racm	Naphthalene aerosol from RO ₂ +HO ₂ reactions
AISO1,2	updated	cb05e51, saprc07tb, saprc07tc*, racm	Aerosol from isoprene reactions NO ₃ added to existing OH (all yields follow the OH pathway)
AISO3	updated	cb05e51, saprc07tb, saprc07tc*, racm	Aerosol from reactive uptake of IEPOX on aqueous aerosol particles. Specifically intended to be the sum of 2-methyltetrols and IEPOX-derived organosulfates.
AALK1,2	added	cb05e51, saprc07tb, saprc07tc, saprc07tic, racm	Alkane aerosol
AALK	removed	all	deprecated alkane aerosol

*AERO6i does not include SOA from isoprene+NO₃ in AISO1,2 (it is included in AISOPNNJ). AERO6i does not include IEPOX SOA in AISO3 (it is included in AITETJ, AIEOSJ, AIDIMJ, etc). AISO3 is approximately zero in AERO6i.

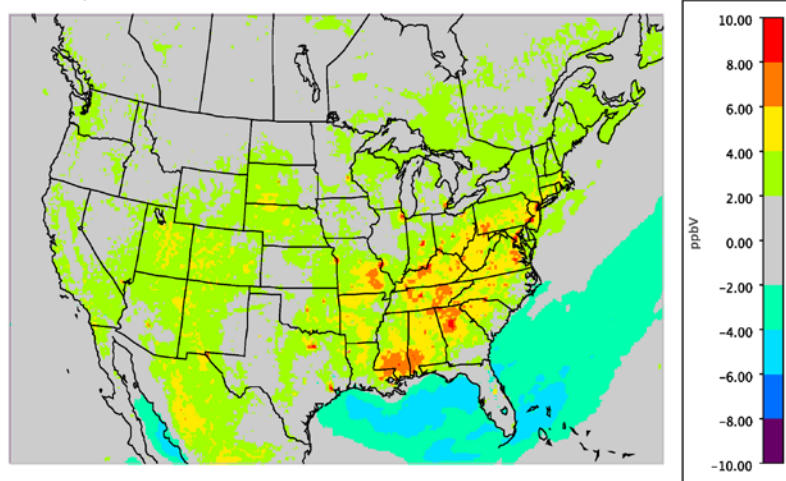
Table 2: Description of the CMAQ model simulations utilized.

CMAQ Simulation Name	CMAQ version	WRF Version	NEI Version	Photolysis Scheme	Chemical Mechanism	Simulation Period
CMAQv5.0.2_Base	v5.0.2	v3.4	v1	v5.0.2	CB05TUCL	Annual (2011)
CMAQv5.0.2_WRFv3.7	v5.0.2	v3.7	v1	v5.0.2	CB05TUCL	January and July 2011
CMAQv5.1_Base	v5.1	v3.7	v2	v5.1	CB05e51	Annual (2011)
CMAQv5.1_RetroPhot	v5.1	v3.7	v2	v5.0.2	CB05e51	January and July 2011
CMAQv5.1_TUCL	v5.1	v3.7	v2	v5.1	CB05TUCL	January and July 2011

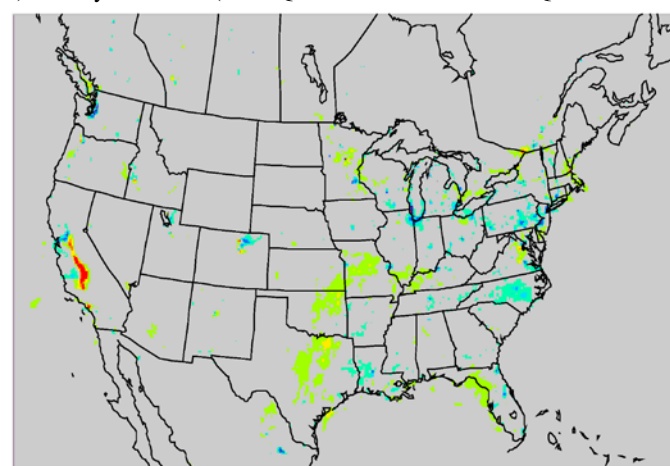
a) January O₃ diff (CMAQv5.0.2_WRFv3.7 – CMAQv5.0.2_Base)



b) July O₃ diff (CMAQv5.0.2_WRFv3.7 – CMAQv5.0.2_Base)



c) January PM_{2.5} diff (CMAQv5.0.2_WRFv3.7 - CMAQv5.0.2_Base)



d) July PM_{2.5} diff (CMAQv5.0.2_WRFv3.7 – CMAQv5.0.2_Base)

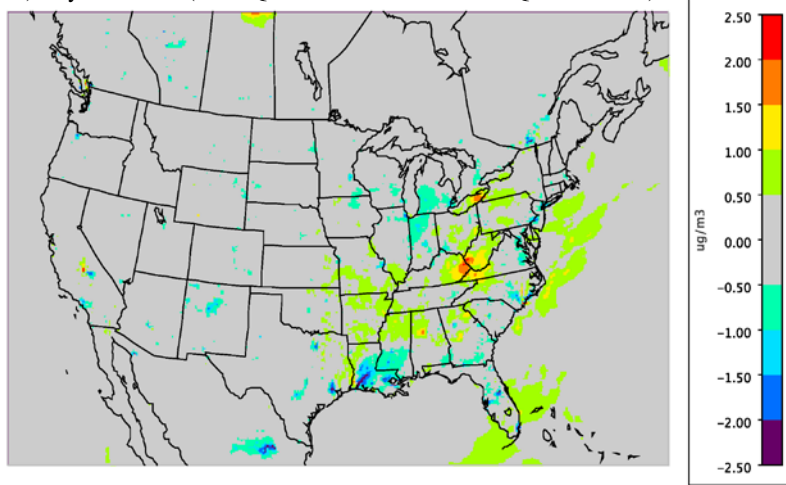
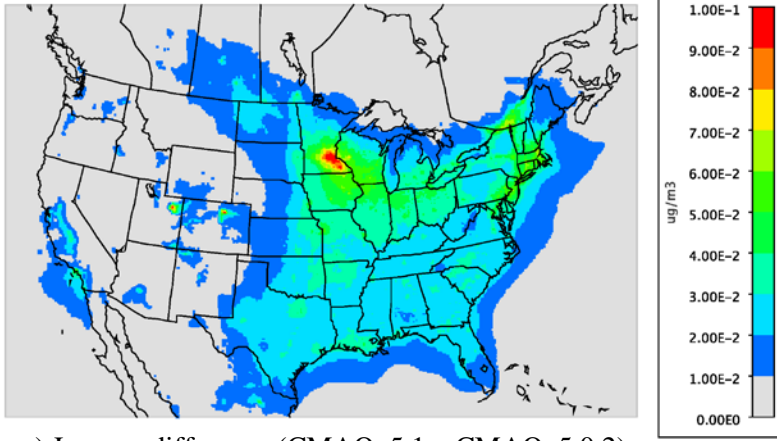
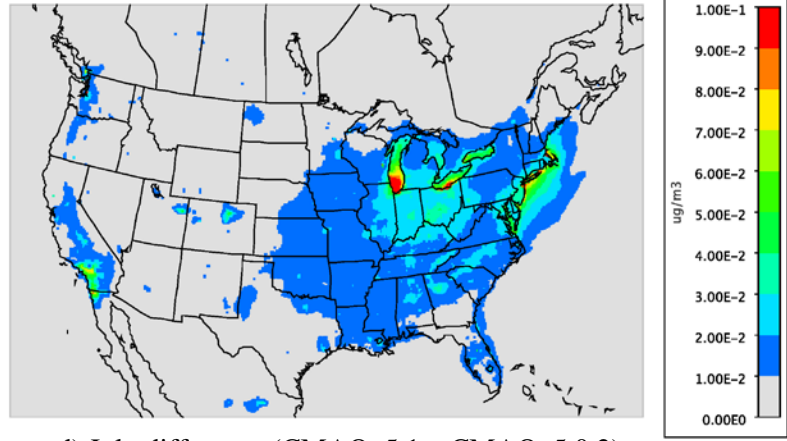


Figure 1: Monthly average difference in O₃ (ppbv) for a) January and b) July and PM_{2.5} (μg m⁻³) for c) January and d) July between CMAQv5.0.2 using WRFv3.4 (CMAQv5.0.2_Base) and CMAQv5.0.2 using WRFv3.7 (CMAQv5.0.2_WRFv3.7) (CMAQv5.0.2_WRFv3.7 – CMAQv5.0.2_Base). Note that the scales for each plot can vary.

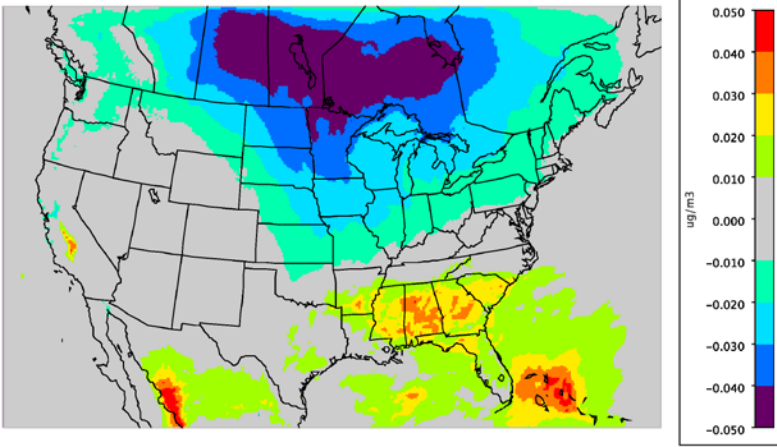
a) January difference (CMAQv5.1 – CMAQv5.0.2)



b) July difference (CMAQv5.1 – CMAQv5.0.2)



c) January difference (CMAQv5.1 – CMAQv5.0.2)



d) July difference (CMAQv5.1 – CMAQv5.0.2)

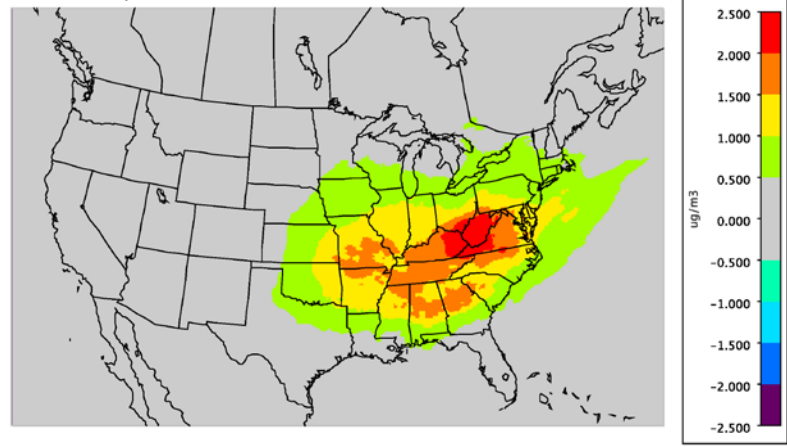
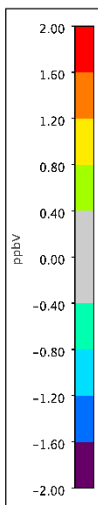
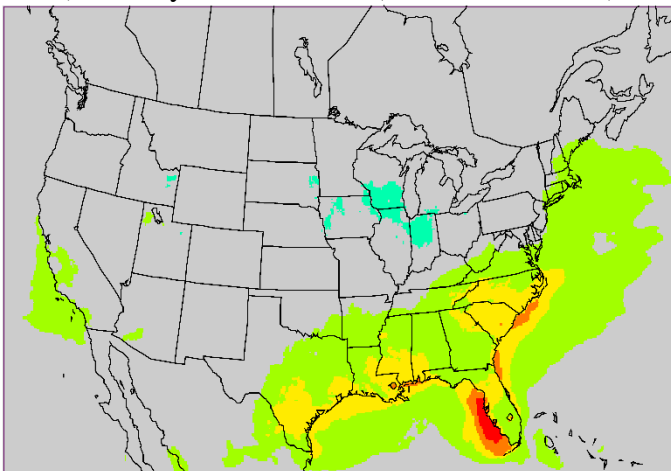
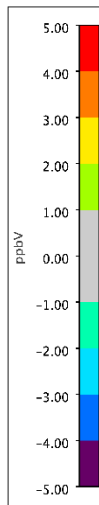
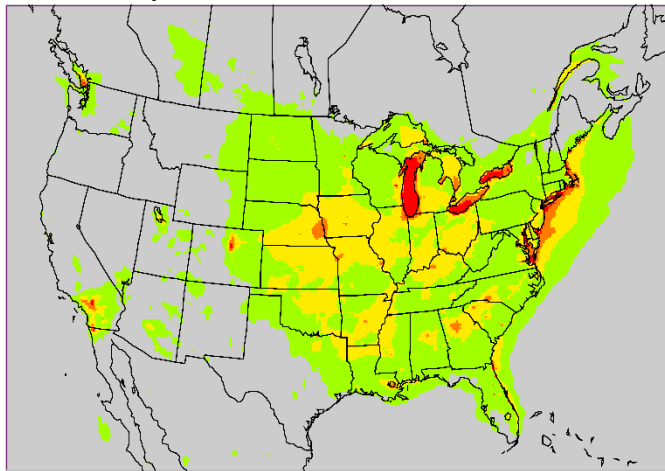


Figure 2: Monthly average sum total of AALK1, AALK2, APAH1, APAH2 and APAH3 for a) January and b) July (upper right) and the monthly average difference is the sum total of AISO1, AISO2, AISO3 and AOLGB for c) January and d) July between the aerosol treatments in CMAQ v5.0.2 and v5.1 (v5.1 – v5.0.2). All plots are in units of $\mu\text{g m}^{-3}$. Note that the scales for each plot can vary.

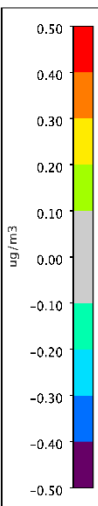
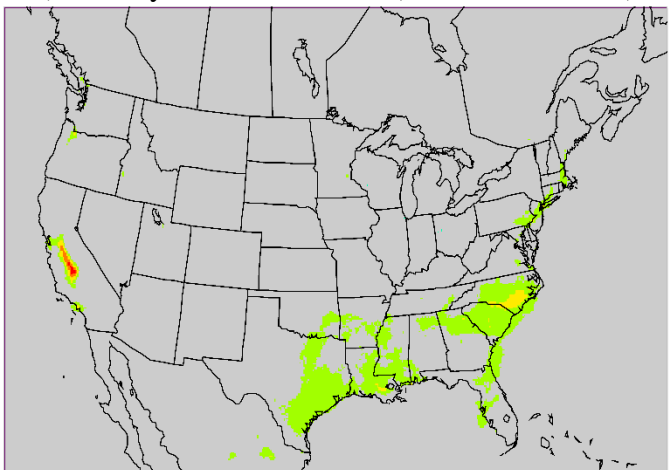
a) January O₃ difference (Base – RetroPhot)



b) July O₃ difference (Base – RetroPhot)



c) January PM_{2.5} difference (Base – RetroPhot)



d) July PM_{2.5} difference (Base – RetroPhot)

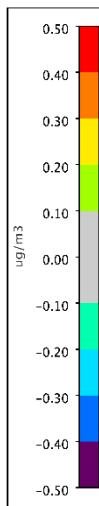
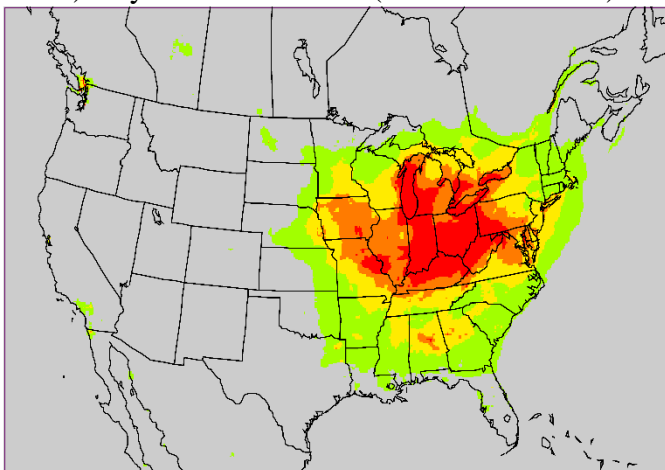


Figure 3: Difference in the monthly average O₃ for a) January and b) July and PM_{2.5} for c) January and d) July between CMAQ v5.1_base and v5.1_RetroPhot (v5.1_Base – v5.1_RetroPhot). O₃ plots are in units of ppb and PM_{2.5} plots are in units of μgm^{-3} . Note that the scales for each plot can vary.

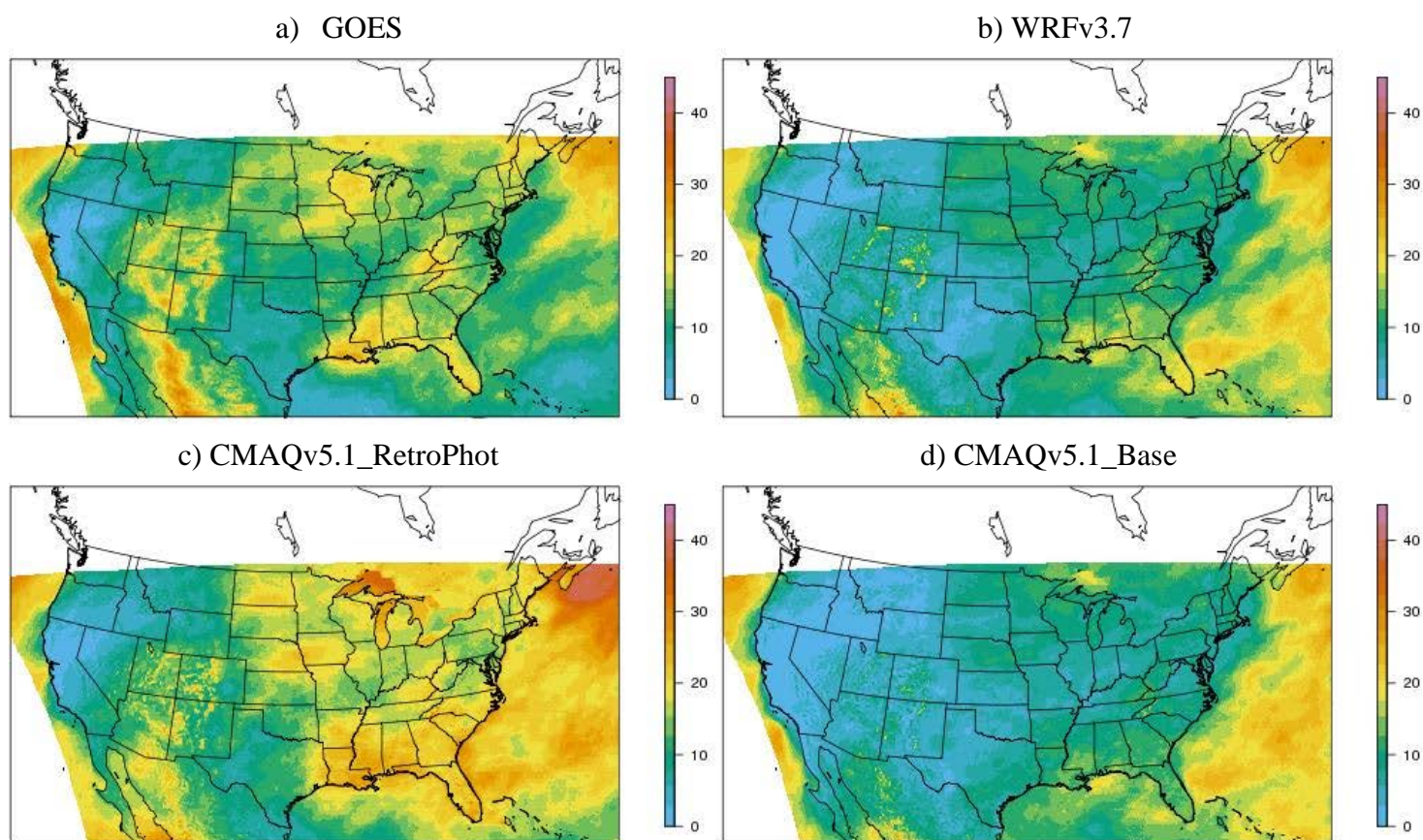


Figure 4. The average cloud albedo during daytime hours in July 2011 derived from (a) the GOES satellite product (b) WRF3.7 (c) CMAQv5.1 with photolysis/cloud model treatment from v5.0.2 and WRF3.7 inputs (CMAQv5.1_RetroPhot) (d) CMAQv5.1 using WRF3.7 inputs (CMAQv5.1_Base).

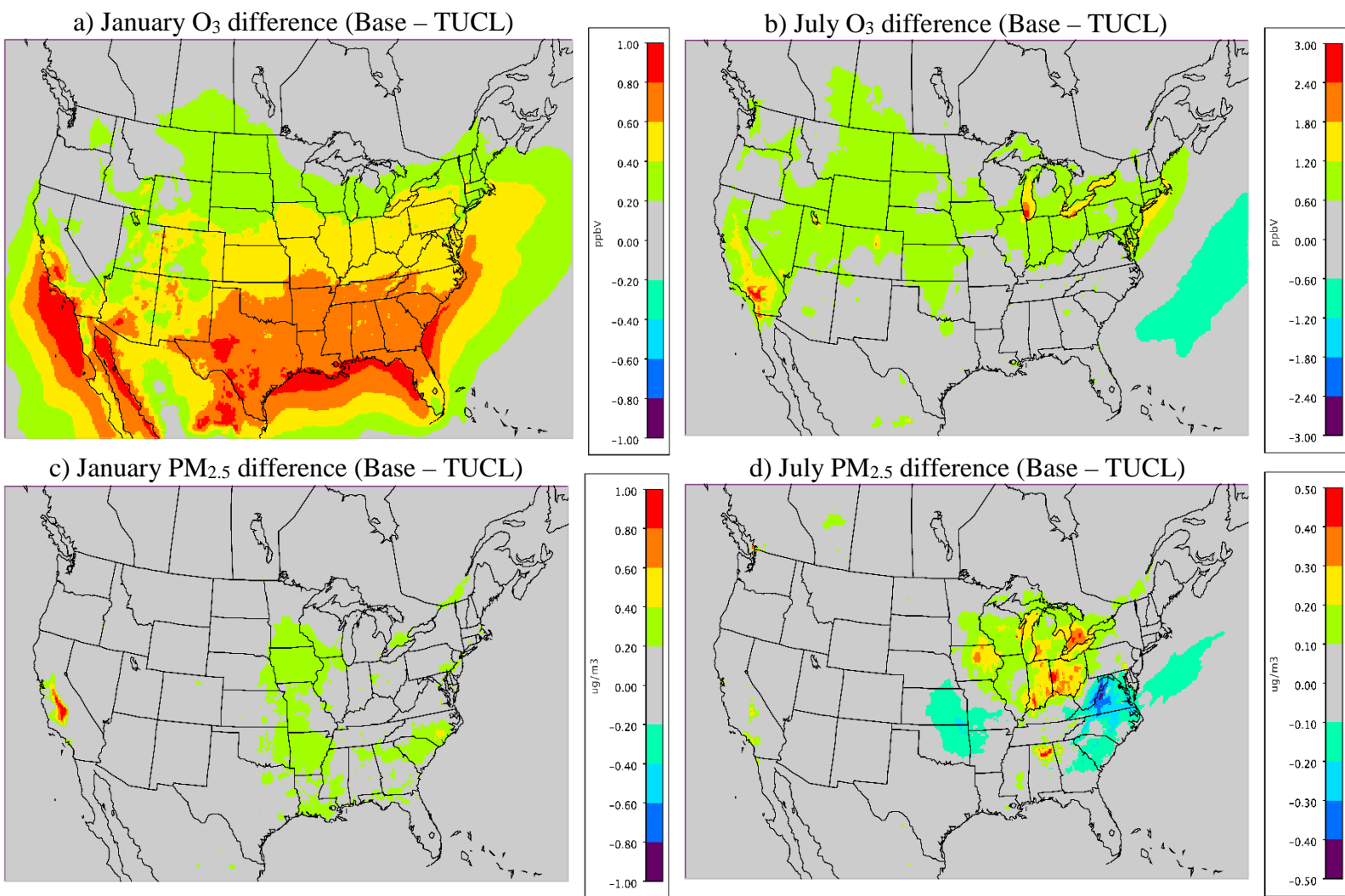


Figure 5: Difference in the monthly average O₃ for a) January and b) July and PM_{2.5} (with organic matter mass removed) for c) January and d) July between CMAQ v5.1_Base and v5.1_TUCL (CMAQv5.1_Base – CMAQv5.1_TUCL). O₃ plots are in units of ppb and PM_{2.5} plots are in units of $\mu\text{g}/\text{m}^3$. Note that the scales for each plot can vary.

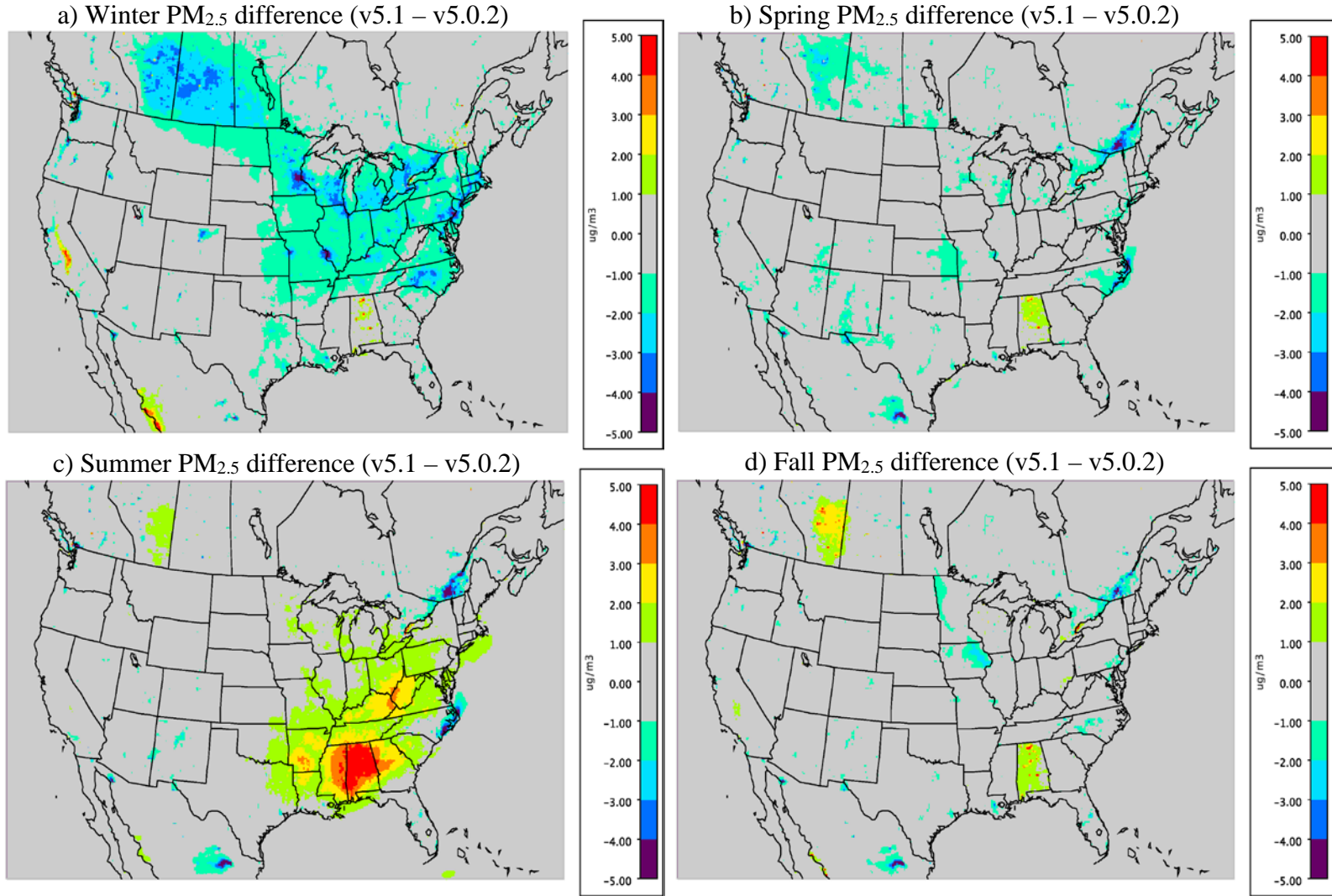


Figure 6: Difference in the seasonal average $PM_{2.5}$ for a) winter (DJF) b) spring (MAM) c) summer (JJA) and d) fall (SON) between CMAQ v5.0.2_Base and v5.1_Base (CMAQv5.1_Base – CMAQv5.0.2_Base). All plots are in units of $\mu g m^{-3}$.

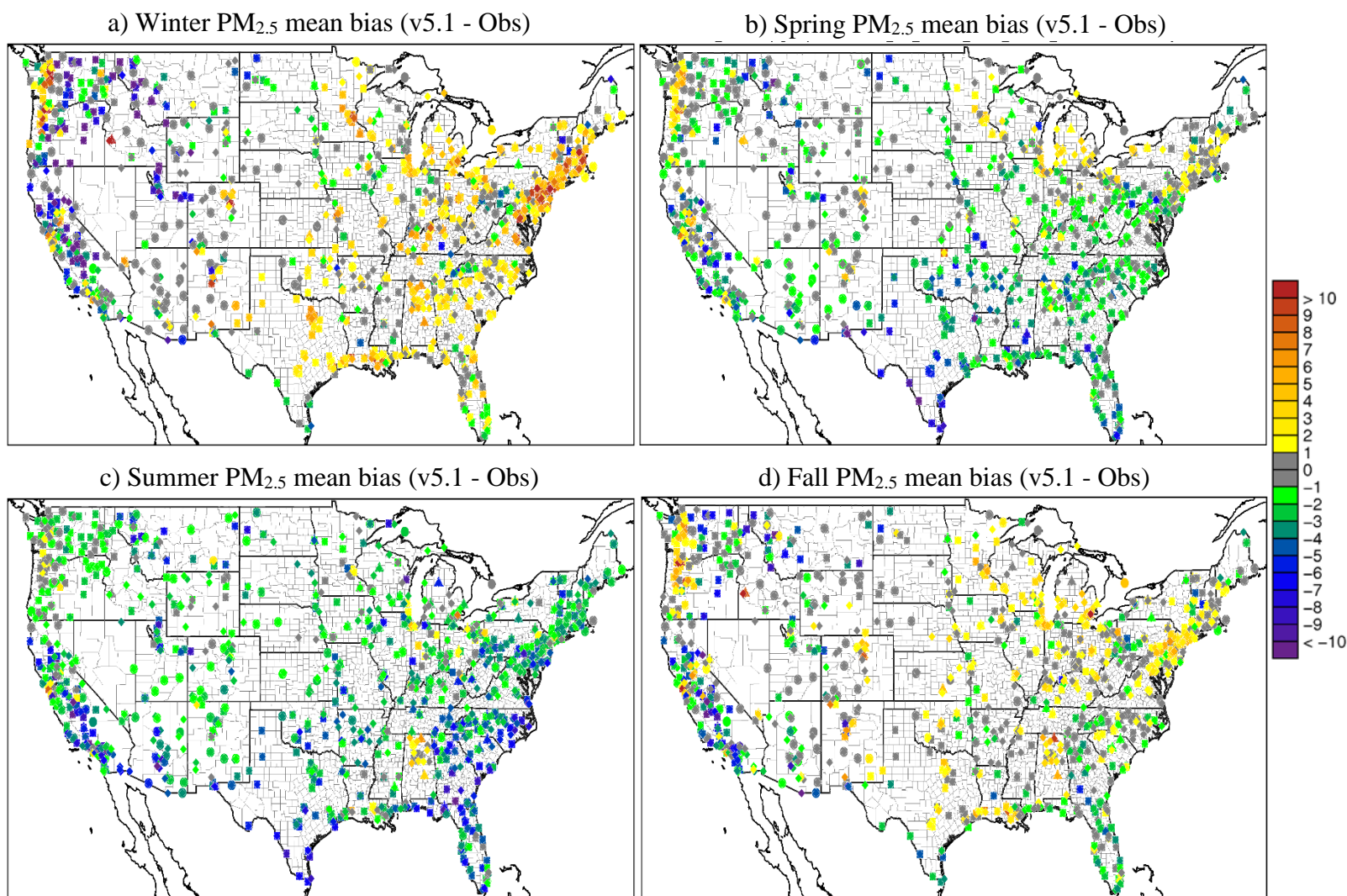


Figure 7: Seasonal average PM_{2.5} mean bias ($\mu\text{g}\text{m}^{-3}$) at IMPROVE (circles), CSN (triangles), AQS Hourly (squares) and AQS Daily (diamonds) sites for a) winter (DJF) b) spring (MAM) c) summer (JJA) and d) fall (SON) for the CMAQ v5.1_Base simulation.

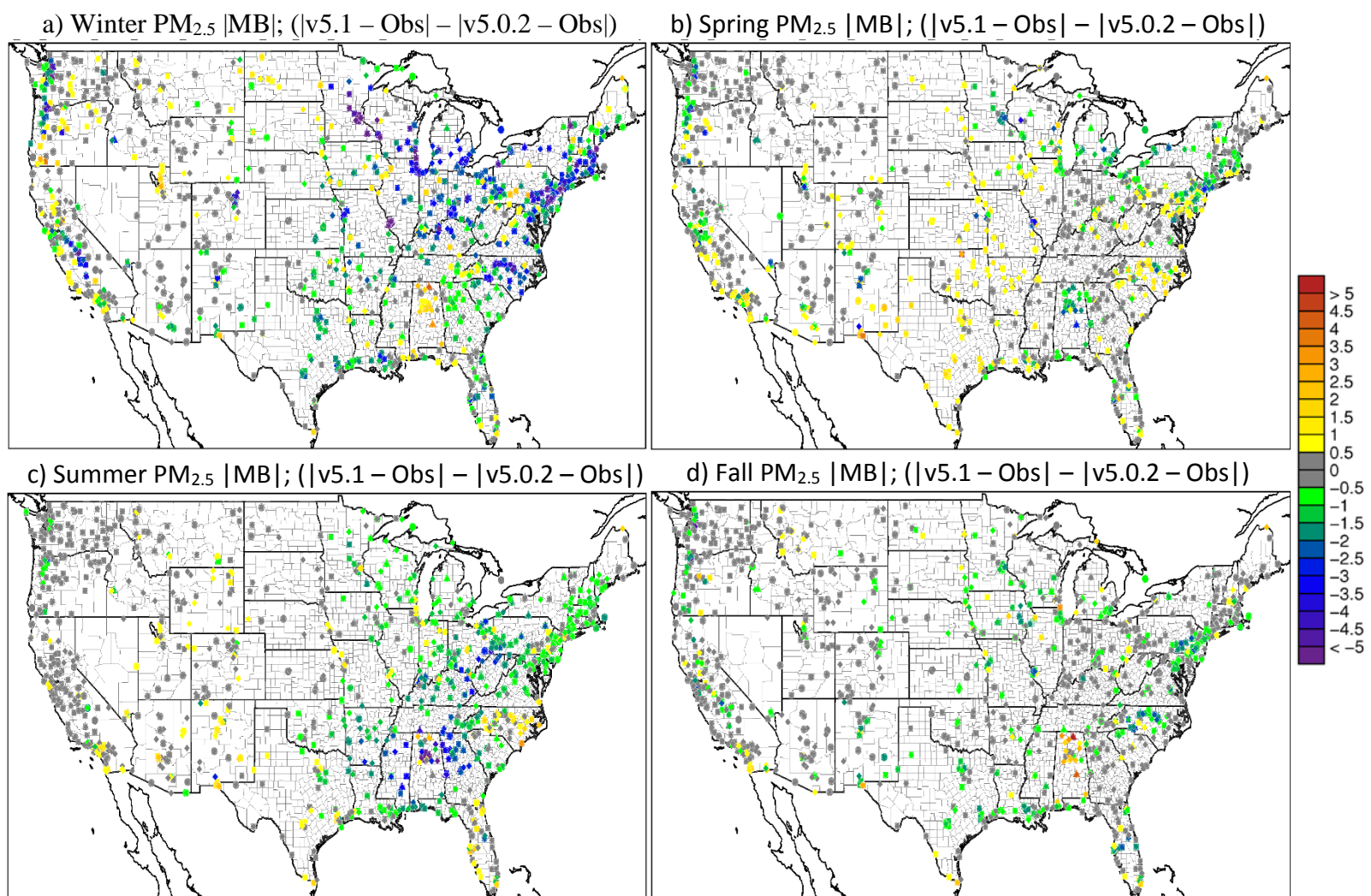


Figure 8: Difference in the absolute value of seasonal average PM_{2.5} mean bias for a) winter (DJF) b) spring (MAM) c) summer (JJA) and d) fall (SON) between CMAQ v5.0.2_Base and v5.1_Base (CMAQv5.1_Base – CMAQv5.0.2_Base). All plots are in units of μgm^{-3} . Cool colors indicate a reduction in PM_{2.5} mean bias in v5.1 while warm color indicate an increase in PM_{2.5} mean bias v5.1.

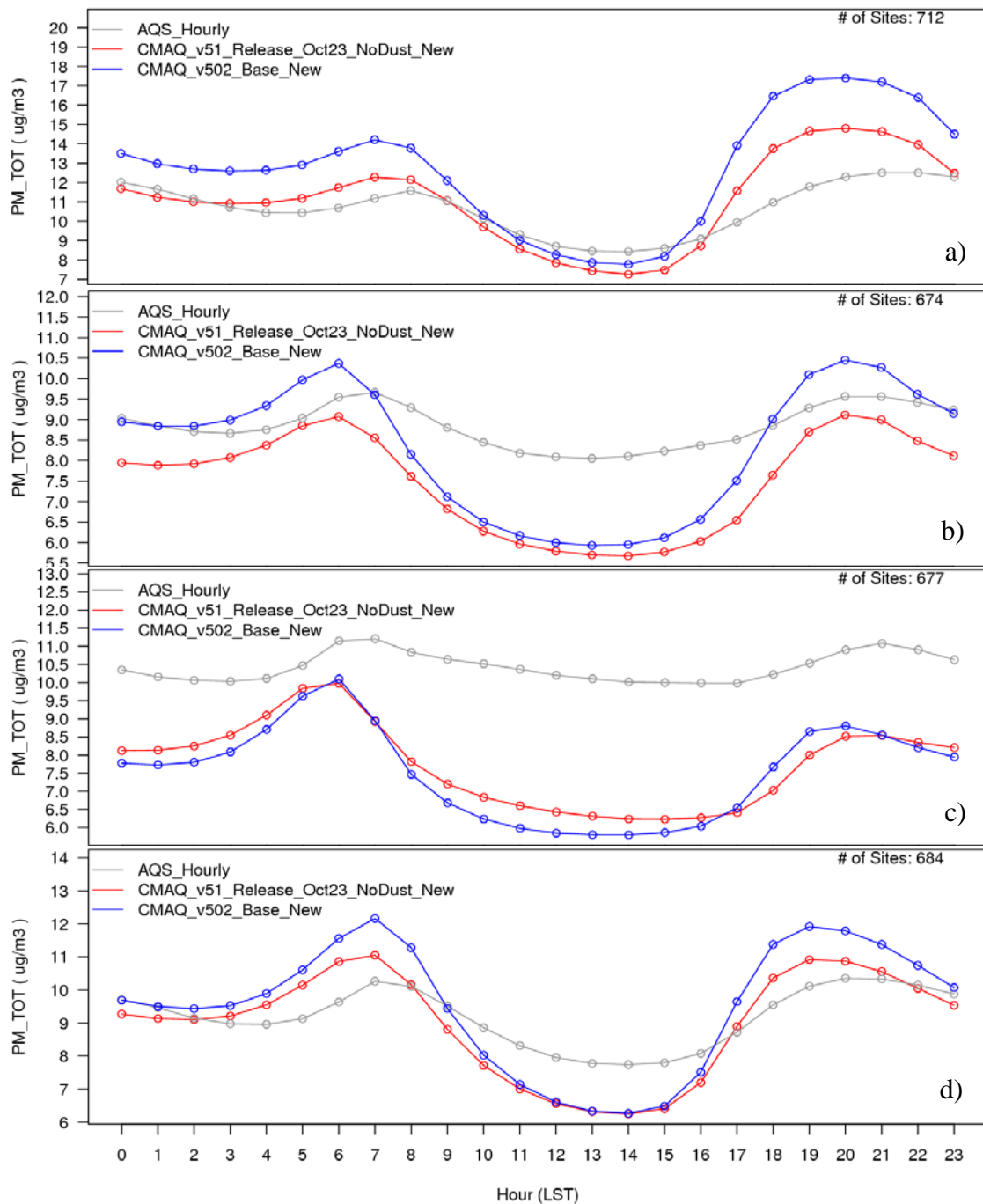


Figure 9: Diurnal time series of seasonal $PM_{2.5}$ ($\mu g/m^3$) from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for a) winter b) spring c) summer and d) fall.

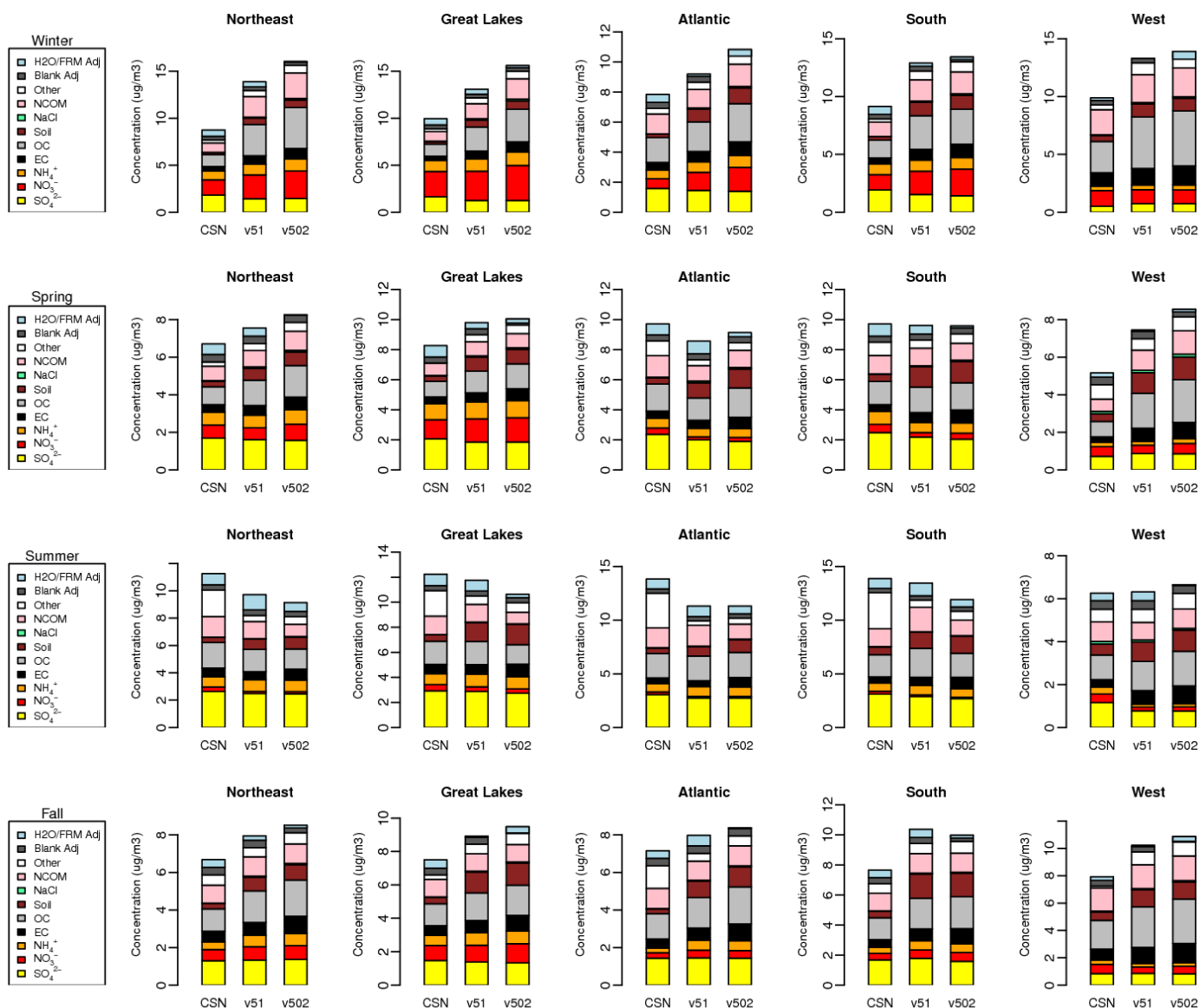
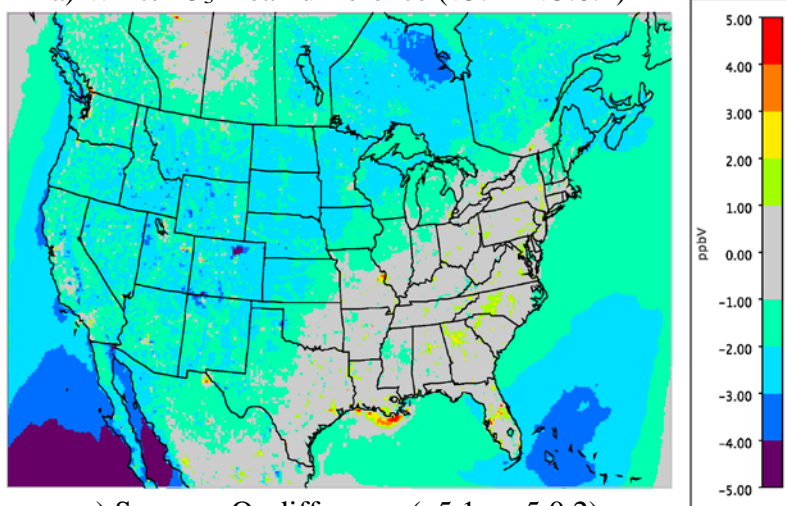
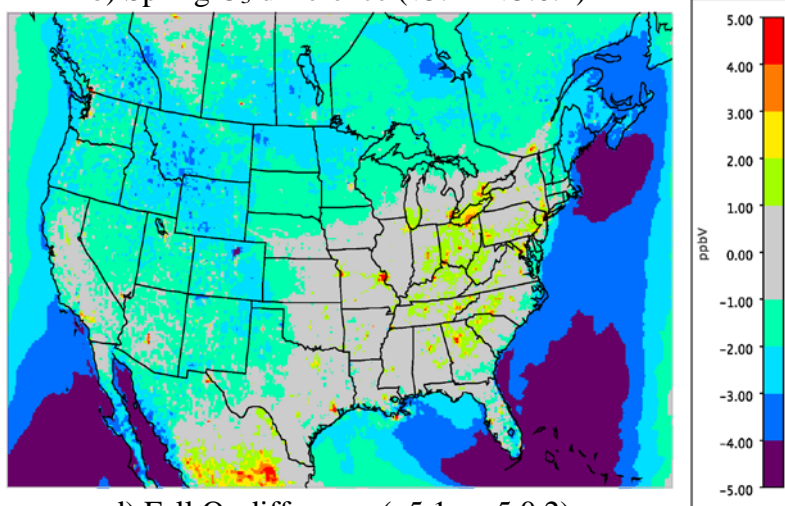


Figure 10: Regional and seasonal stacked bar plots of PM_{2.5} composition at CSN sites. In order from top to bottom are spring, summer, fall and winter seasons and left to right the Northeast, Great Lakes, Atlantic, South and West regions. The individual PM_{2.5} components (in order from bottom to top) are SO₄²⁻ (yellow), NO₃⁻ (red), NH₄⁺ (orange), EC (black), OC (light gray), Soil (brown), NaCl (green), NCOM (pink), other (white), blank adjustment (dark gray) and H₂O/FRM adjustment (blue).

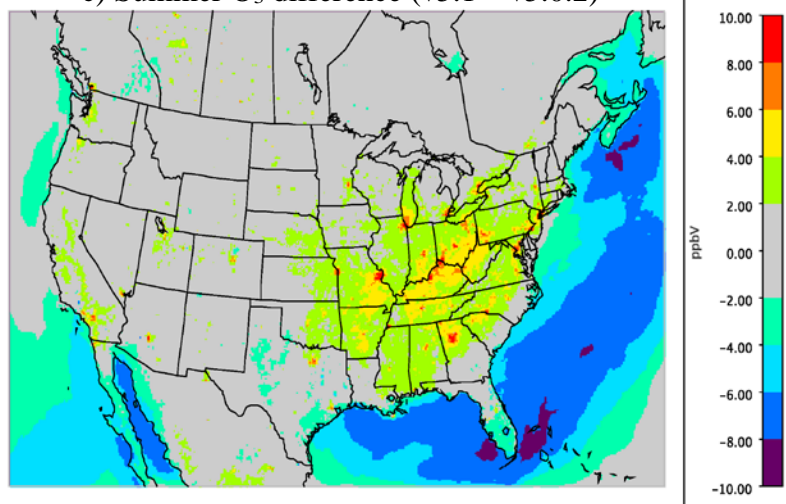
a) Winter O₃ mean difference (v5.1 – v5.0.2)



b) Spring O₃ difference (v5.1 – v5.0.2)



c) Summer O₃ difference (v5.1 – v5.0.2)



d) Fall O₃ difference (v5.1 – v5.0.2)

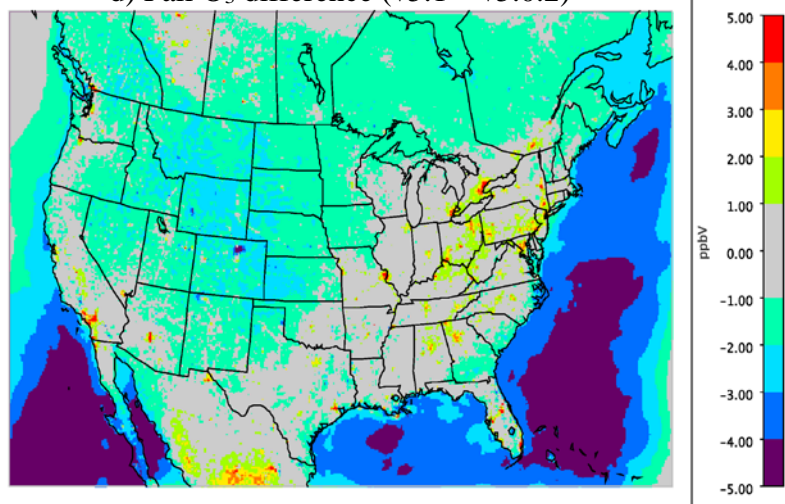


Figure 11: Difference in the monthly average hourly O₃ (ppbv) for winter (DJF; top left), spring (MAM; top right), summer (JJA; bottom left) and fall (SON; bottom right) between CMAQ v5.0.2_Base and v5.1_Base (CMAQv5.1_Base – CMAQv5.0.2_Base). Note that the scales for each plot can vary.

a) Winter O₃ mean bias (v5.1 - Obs)

b) Spring O₃ mean bias (v5.1 - Obs)

c) Summer O₃ mean bias (v5.1 - Obs)

d) Fall O₃ mean bias (v5.1 - Obs)

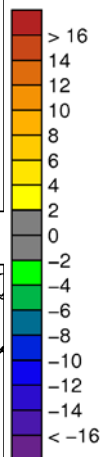


Figure 12: Seasonal average hourly O₃ mean bias at AQS sites for a) winter (DJF) b) spring (MAM) c) summer (JJA) and d) fall (SON) for the CMAQ v5.1_Base simulation. All plots are in units of ppbV.

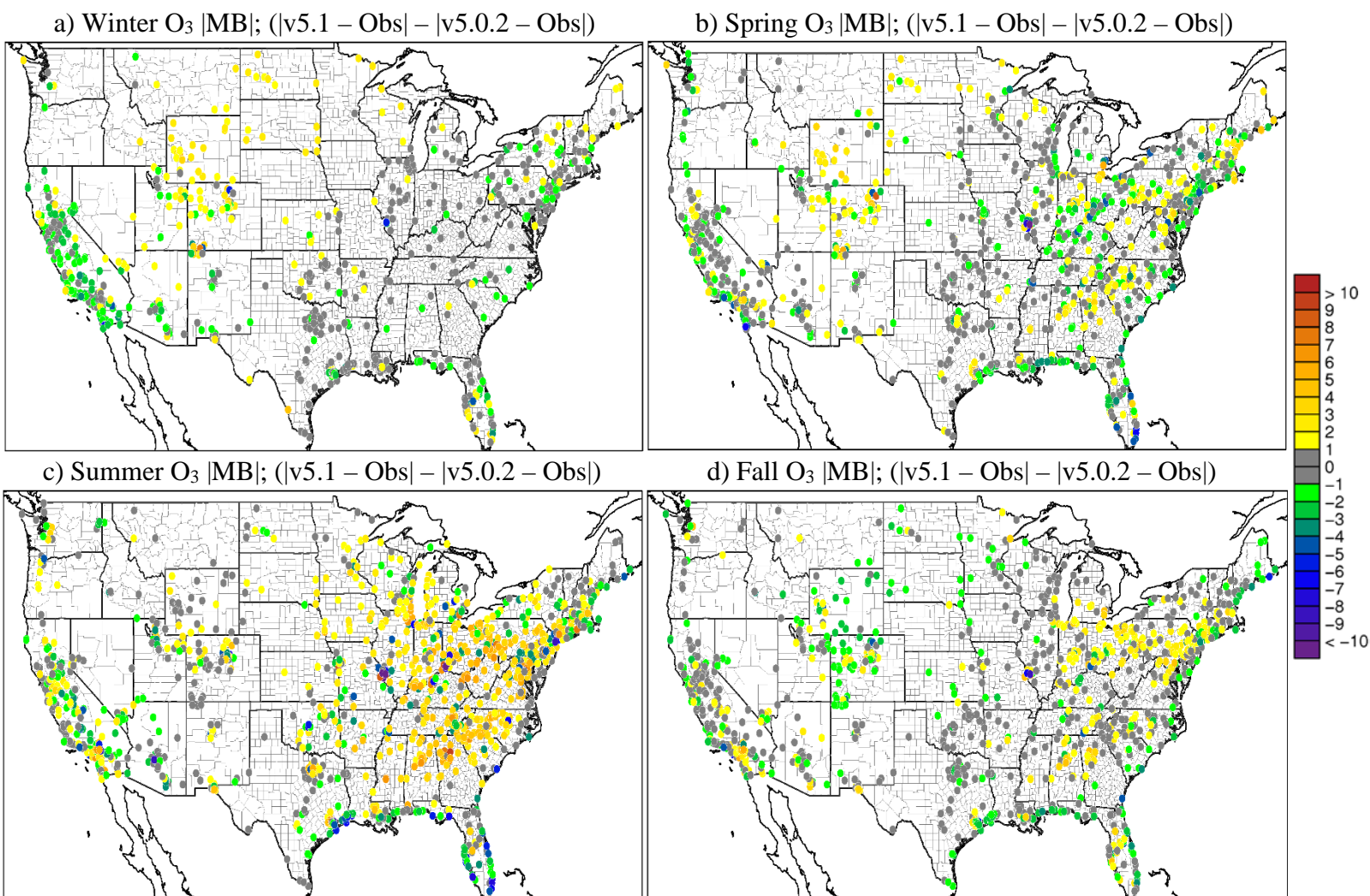


Figure 13: Difference in the absolute value of monthly average O₃ mean bias for a) winter (DJF) b) spring (MAM) c) summer (JJA) and d) fall (SON) between CMAQ v5.0.2_Base and v5.1_Base (CMAQv5.1_Base – CMAQv5.0.2_Base). All plots are in units of ppbV. Cool colors indicate a reduction in O₃ mean bias in v5.1 while warm color indicate an increase in O₃ mean bias v5.1.

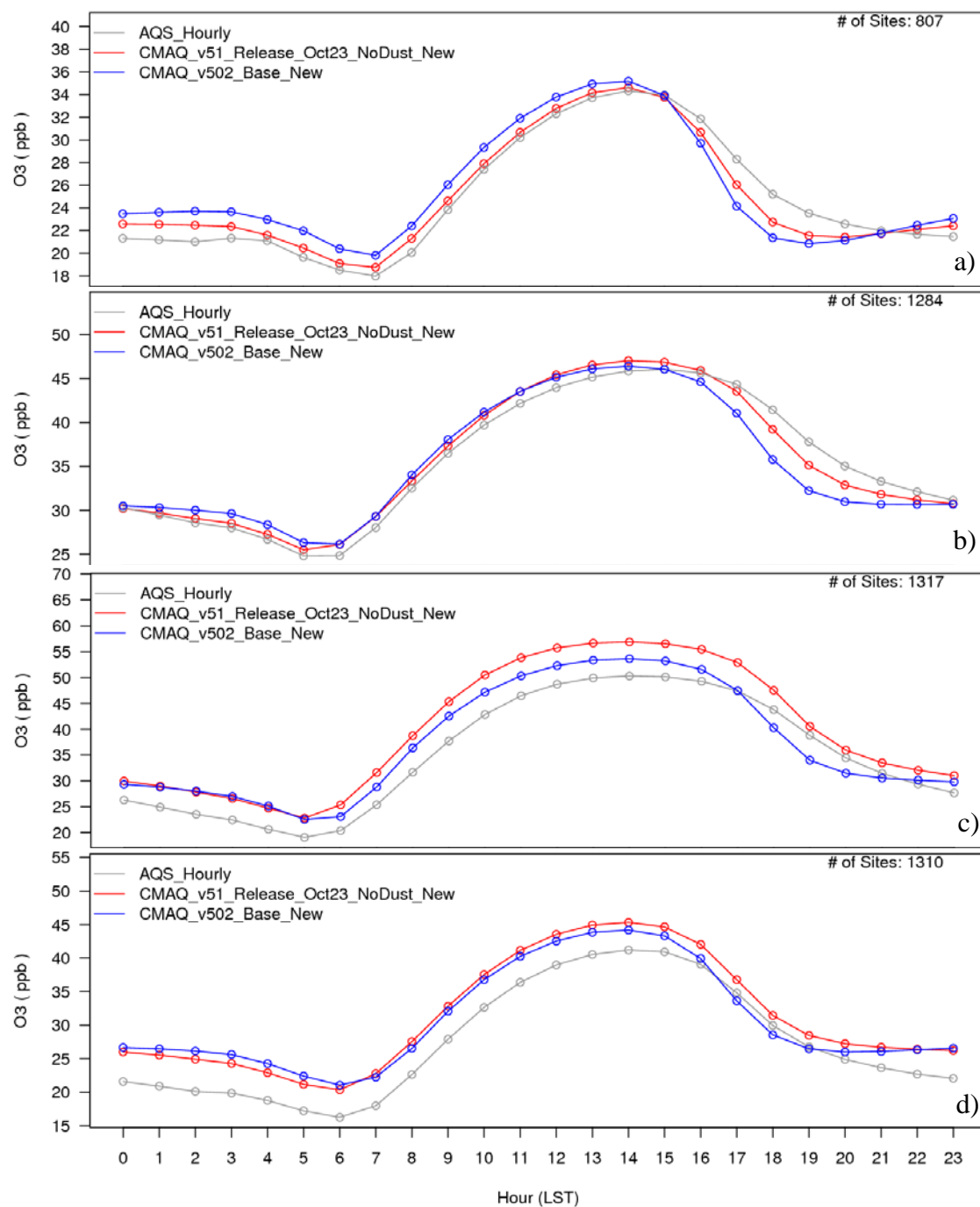


Figure 14: Diurnal time series of seasonal O₃ (ppbv) from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for a) winter b) spring c) summer and d) fall.

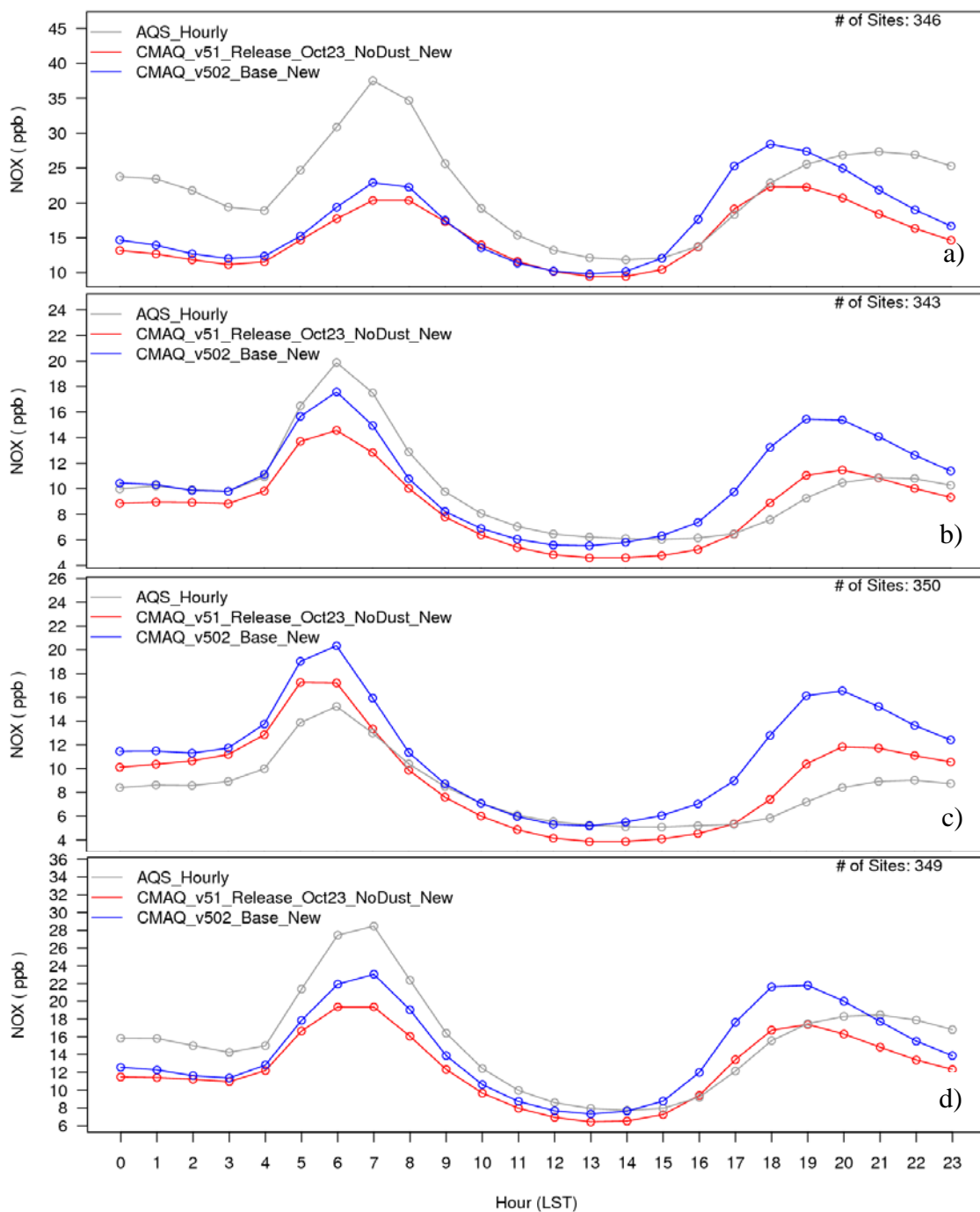


Figure 15: Diurnal time series of seasonal NO_x (ppbv) from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for a) winter b) spring c) summer and d) fall.

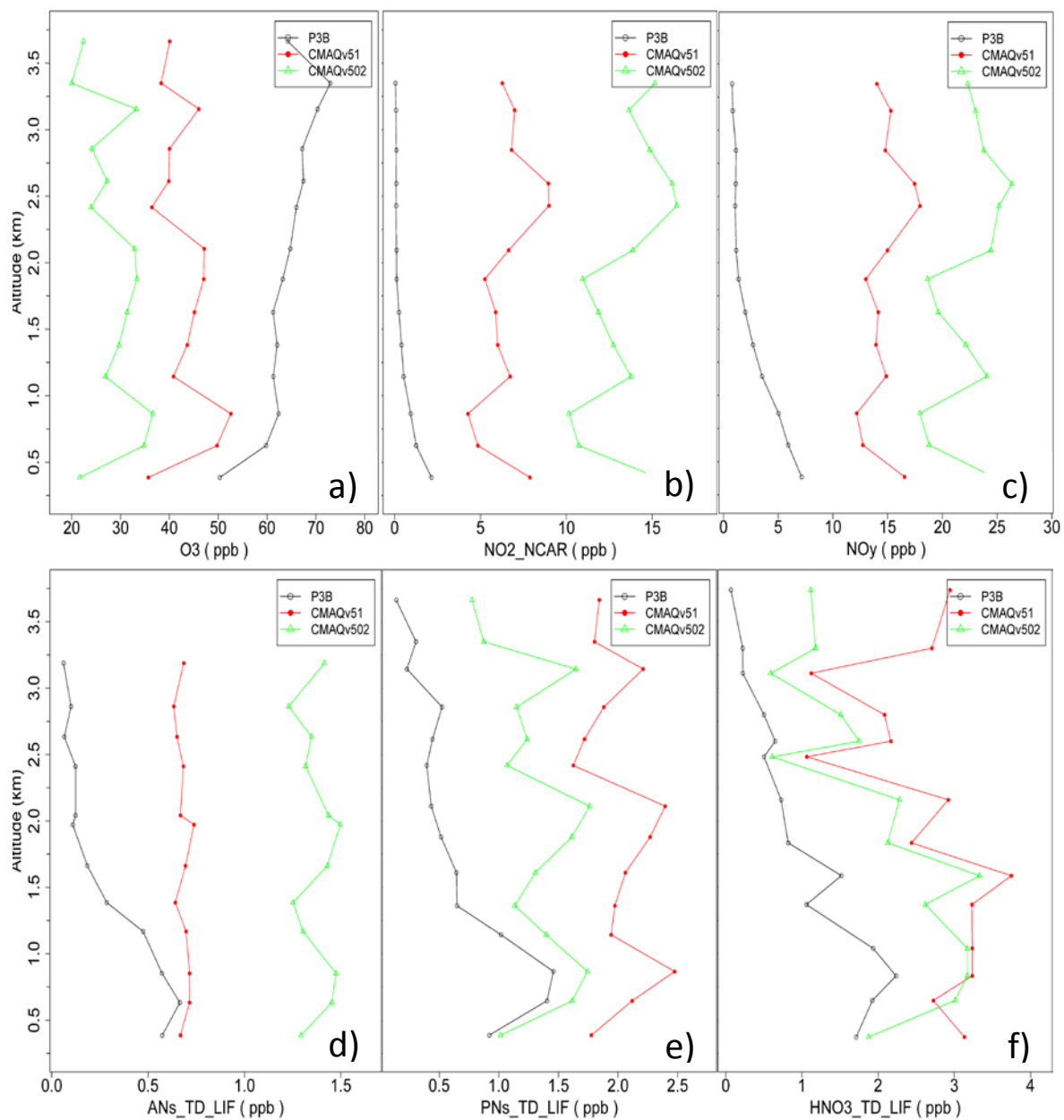


Figure 16: Observed (black) and CMAQ simulated vertical profiles of a) O_3 b) NO_2 c) NO_y d) alkyl nitrates (ANs) e) peroxy nitrates (PNs) and f) HNO_3 for the Edgewood site in Baltimore, MD on July 5, 2011. CMAQv502_Base simulation profiles are shown in green and CMAQv51_Base simulation profiles are shown in red. Altitude (km) is given on the y-axis, while mixing ratio (ppbv) is given on the x-axis.

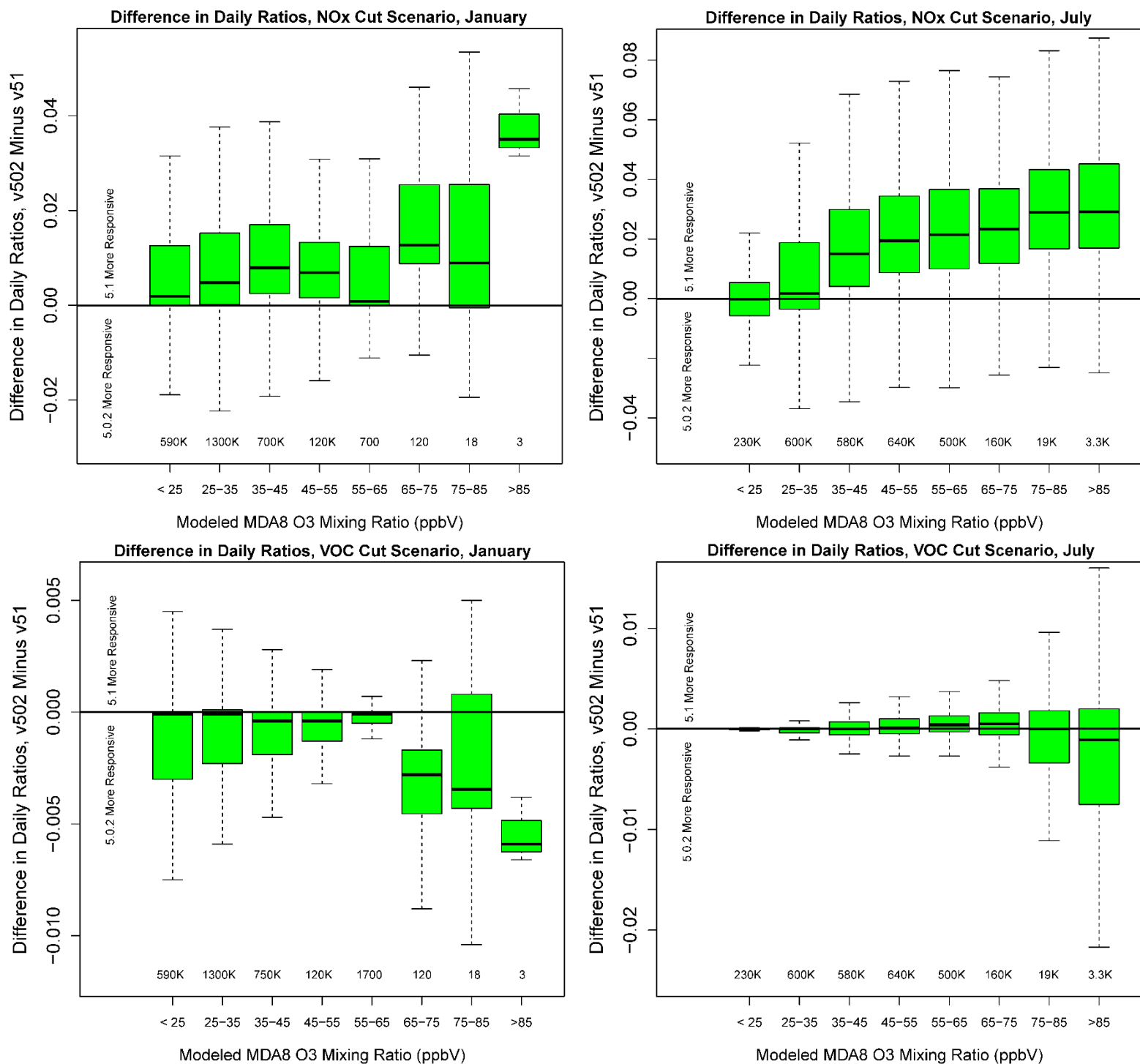


Figure 17: Difference in MDA8 O₃ daily ratios (Cut Scenario / Base) for CMAQv502 and v51 (v502 – v51) for a 50% cut in anthropogenic NO_x (top) and VOC (bottom) for January (left) and July (right) binned by the modeled MDA8 O₃ mixing ratio (ppbV). Values greater than one indicate v51 is more responsive than v502 to the emissions cut, while values less than one indicate v502 is more responsive. Given above the x-axis is the number of model grid cells in each bin.

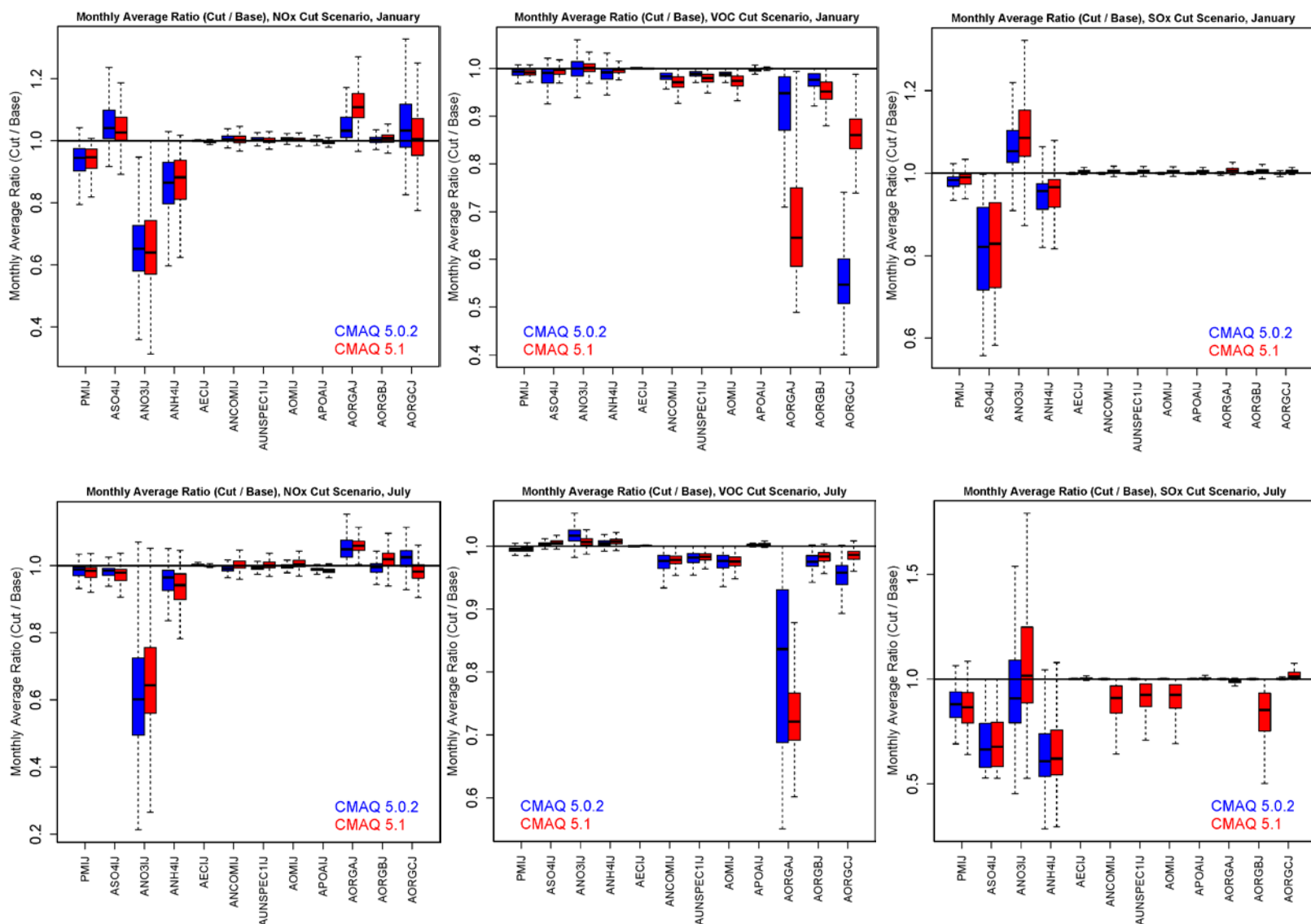
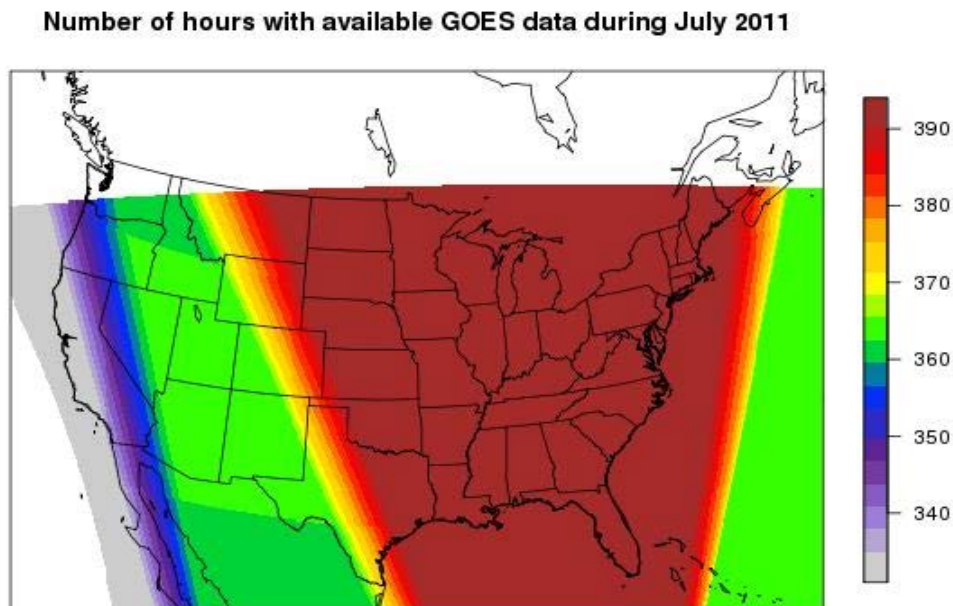


Figure 18: Box plots of monthly average ratio values (Cut / Base) of PM₁₀ (total PM_{2.5}), ASO₄, ANO₃, ANH₄, AECI, ANCOMI, AUNSPECI, AOMI, APOAI, AORGAJ, AORGBJ, and AORGCJ for v502 (blue) and v51 (red) for a 50% cut in anthropogenic NO_x (left), VOC (middle) and SO_x (right) for January (top) and July (bottom).

Section S.1: Use of satellite measured cloud albedo to evaluate cloud parameterizations in CMAQ and WRF

Cloud albedo is a measure of the solar radiation that is reflected by a cloud and is one of the Imager products available from NASA's Geostationary Operational Environmental Satellite (GOES). The figure below shows the number of daytime hours (11:45UTC – 23:45UTC) with available GOES cloud albedo data during July 1 – July 31, 2011 for the modeling domain.



Although the WRF and CMAQ systems do not use cloud albedo directly in their cloud parameterizations, this variable provides a useful model diagnostic for identifying areas where the models are over or under-predicting the degree of cloudiness over a region.

WRF cloud albedo is calculated as:

$$\text{CLDALB_WRF} = (\text{SWUPT} - \text{SWUPTC}) / \text{SWDNT} * 100\% \quad (1)$$

where SWUPT is the upwelling shortwave flux, SWUPTC is the upwelling clear sky shortwave flux and SWDNT is the downwelling shortwave flux. All three fluxes are instantaneous and at the top of the model.

Cloud albedo within the CMAQv5.1 and CMAQv5.0.2 photolysis module is calculated as:

$$\text{CLDALB_NEW} = \pi * (\text{REFLECTION} - \text{CLR_REFLECTION}) \quad (2)$$

where REFLECTION is the shortwave reflection, CLR_REFLECTION is the clear sky shortwave reflection, both are instantaneous and at the top of the model.

Table S1 provides categorical evaluation metrics for daytime hours (11:45UTC – 23:45UTC) in July 1 – July 31, 2011 for model predicted clouds based on the CMAQv5.0.2 photolysis module, the CMAQv5.1 photolysis module and WRFv3.7 cloud parameterization compared to GOES satellite data. Cloud albedo is used to determine the presence of clouds in each grid cell for each hour. A cloud albedo > 5% (modeled or observed) is used as the threshold to indicate cloudy conditions. The Agreement Index (Biazer et al., 2014) is the fraction of correct model predictions (true clear skies OR true cloudy conditions) out of all grid cell/hours with available GOES data. Model over-predictions (3rd row) are the number of grid cell/hours where the model predicted cloudy conditions but the GOES product showed clear skies. Model under-predictions indicate the model predicted clear skies when the GOES product show cloudy conditions. Each metric is calculated separately for all hourly data available at land grid cells (N=25,771,567) and all hourly data available at ocean grid cells (N=12,376,594).

The relative frequency of model over-prediction of cloudy conditions over land decreased from 0.22 in CMAQv5.0.2 to 0.07 in CMAQv5.1 and is now consistent with the WRF evaluation over land. However, CMAQv5.1 over-predicts clouds over the ocean to a greater extent than either CMAQv5.0.2 or WRF3.7 (i.e. the over prediction relative frequency is 0.33 compared to 0.27 or 0.21). This issue will be addressed by new science updates in the CMAQ system and evaluation results are expected to improve in upcoming CMAQ releases. WRFv3.7 and CMAQv5.1 under-predict cloudy conditions over land 26% of the time. Resolving this issue will require changes to the WRF cloud parameterization.

Table S1.

Categorical Metric	CMAQv5.0.2 Photolysis		CMAQv5.1 Photolysis		WRFv3.7	
	Land	Ocean	Land	Ocean	Land	Ocean
Agreement Index	0.66	0.61	0.67	0.58	0.67	0.66
Over-predict clouds	0.22	0.27	0.07	0.33	0.07	0.21
Under-predict clouds	0.12	0.12	0.26	0.09	0.26	0.13

Reference:

Biazar, A.P., 2014: Cloud Correction and its Impact on Air Quality Simulations, presented at the 94th AMS Annual Meeting, Atlanta, GA.

S.2 CMAQv5.1 CB05e51 AE6 Species Definitions (available with the CMAQv5.1 release code)

! Updated AOCIJ, AOMIJ, and AORGAJ definition for CMAQv5.1 based on recommendations from Havala Pye (Dec 2014)

! The formulas used in this file implicitly assume that the model-ready emission files were prepared using a GSPRO from the 2002 emissions platform or later, in which POC emissions (hence, the CMAQ species APOCI and APOCJ) represent pure organic carbon without any scaling factor for OM:OC ratios.

! Output variables that begin with 'PM' represent those in which a size cut was applied. For example, PM25_NA is all sodium that falls below 2.5 um diameter. These 'PM' variables are used for comparisons at IMPROVE and STN sites.

! Output variables beginning with 'A' (aside from AIR_DENS) represent a combination of aerosol species in which no size cut was applied. For example, ASO4IJ is the sum of i-mode and j-mode sulfate. These 'A' variables are used for comparisons at CASTNet sites.

! Output variables beginning with 'PMC' is the coarse mode of total PM, i.e., sums all modes then subtracts the fine mode (PM2.5). These 'PMC' variables are used for comparisons at SEARCH sites.

/ File [1]: CMAQ conc/aconc file

/ File [2]: AEROVIS file

/ File [3]: METCRO3D file

/ File [4]: AERODIAM file

/ File [5]: METCRO2D file

/

/new species ,units ,expression

! Gases

ALD2 ,ppbV ,1000.0*ALD2[1]

ALDX ,ppbV ,1000.0*ALDX[1]

BENZENE ,ppbV ,1000.0*BENZENE[1]

CO ,ppbV ,1000.0*CO[1]

ETH ,ppbV ,1000.0*ETH[1]

ETHA ,ppbV ,1000.0*ETHA[1]

FORM ,ppbV ,1000.0*FORM[1]

H2O2 ,ppbV ,1000.0*H2O2[1]

HNO3 ,ppbV ,1000.0*HNO3[1]

HNO3_UGM3 ,ug/m3 ,1000.0*(HNO3[1]*2.1756*DENS[3])

HONO ,ppbV ,1000.0*HONO[1]

CLNO2 ,ppbV ,1000.0*CLNO2[1]

HOX ,ppbV ,1000.0*(OH[1]+HO2[1])

OH ,ppbV ,1000.0*(OH[1])

IOLE ,ppbV ,1000.0*IOLE[1]

ISOP ,ppbV ,1000.0*ISOP[1]

N2O5 ,ppbV ,1000.0*N2O5[1]

NH3 ,ppbV ,1000.0*NH3[1]

NH3_UGM3 ,ug/m3 ,1000.0*(NH3[1]*0.5880*DENS[3])

NHX ,ug/m3 ,1000.0*(NH3[1]*0.5880*DENS[3])+ANH4I[1]+ANH4J[1]+ANH4K[1]

NO ,ppbV ,1000.0*NO[1]

NO2 ,ppbV ,1000.0*NO2[1]

ANO3_PPB ,ppbV ,(ANO3I[1]+ANO3J[1]+ANO3K[1])/(DENS[3]*(62.0/28.97))

NTR ,ppbV ,1000.0*(NTRON[1]+NTRALK[1]+NTRCN[1]+NTRCNOH[1]+NTRM[1]+NTRI[1]+NTRPX[1])

PANS ,ppbV ,1000.0*(PAN[1]+PANX[1]+OPAN[1]+MAPAN[1])

NOY ,ppbV

,1000.0*(NO[1]+NO2[1]+NO3[1]+2*N2O5[1]+HONO[1]+HNO3[1]+PNA[1]+CRON[1]+CRNO[1]+CRN2[1]+CRPX[1]+CLNO2[1])+PANS[0]+NTR[0]+ANO3_PPB[0]

O3 ,ppbV ,1000.0*O3[1]

OLE ,ppbV ,1000.0*OLE[1]

PAR ,ppbV ,1000.0*PAR[1]

PAN ,ppbV ,1000.0*PAN[1]

PANX ,ppbV ,1000.0*PANX[1]

SO2 ,ppbV ,1000.0*SO2[1]
 SO2_UGM3 ,ug/m3 ,1000.0*(SO2[1]*2.2118*DENS[3])
 SULF ,ppbV ,1000.0*SULF[1]
 TERP ,ppbV ,1000.0*TERP[1]
 TOL ,ppbV ,1000.0*TOL[1]
 VOC ,ppbC
 ,1000.0*(PAR[1]+2.0*ETH[1]+2.0*ETOH[1]+2.0*OLE[1]+7.0*TOL[1]+8.0*XYLMN[1]+FORM[1]+2.0*ALD2[1]+5.0*ISOP
 [1]+2.0*ETHA[1]+4.0*IOLE[1]+2.0*ALDX[1]+10.0*TERP[1]+10.0*NAPH[1])
 XYLMN ,ppbV ,1000.0*XYLMN[1]

! Particles

!! crustal elements

AFEJ ,ug/m3 ,AFEJ[1]
 AALJ ,ug/m3 ,AALJ[1]
 ASIJ ,ug/m3 ,ASIJ[1]
 ATIJ ,ug/m3 ,ATIJ[1]
 ACAJ ,ug/m3 ,ACAJ[1]
 AMGJ ,ug/m3 ,AMGJ[1]
 AKJ ,ug/m3 ,AKJ[1]
 AMNJ ,ug/m3 ,AMNJ[1]
 ASOILJ ,ug/m3 ,2.20*AALJ[1]+2.49*ASIJ[1]+1.63*ACAJ[1]+2.42*AFEJ[1]+1.94*ATIJ[1]

!! other PM species

AHPLUSIJ ,ug/m3 ,(AH3OPI[1]+AH3OPJ[1])*1.0/19.0
 ANAK ,ug/m3 ,0.8373*ASEACAT[1]+0.0626*ASOIL[1]+0.0023*ACORS[1]
 AMGK ,ug/m3 ,0.0997*ASEACAT[1] +0.0032*ACORS[1]
 AKK ,ug/m3 ,0.0310*ASEACAT[1]+0.0242*ASOIL[1]+0.0176*ACORS[1]
 ACAK ,ug/m3 ,0.0320*ASEACAT[1]+0.0838*ASOIL[1]+0.0562*ACORS[1]
 ACLIJ ,ug/m3 ,ACLI[1]+ACLJ[1]
 AECIJ ,ug/m3 ,AECI[1]+AECJ[1]
 ANAIJ ,ug/m3 ,ANAJ[1]+ANAI[1]
 ANO3IJ ,ug/m3 ,ANO3I[1]+ANO3J[1]
 ANO3K ,ug/m3 ,ANO3K[1]
 TNO3 ,ug/m3 ,2175.6*(HNO3[1]*DENS[3])+ANO3I[1]+ANO3J[1]+ANO3K[1]
 ANH4IJ ,ug/m3 ,ANH4I[1]+ANH4J[1]
 ANH4K ,ug/m3 ,ANH4K[1]
 AOIJ ,ugC/m3
 ,(AXYL1J[1]+AXYL2J[1]+AXYL3J[1])/2.0+(ATOL1J[1]+ATOL2J[1]+ATOL3J[1])/2.0+(ABNZ1J[1]+ABNZ2J[1]+ABNZ3J[1])/2.0
 +(AISO1J[1]+AISO2J[1])/1.6+AISO3J[1]/2.7+(ATRP1J[1]+ATRP2J[1])/1.4+ASQTJ[1]/2.1+AALK1J[1]/1.17+AALK2J[1]/1.1
 7+AORGCJ[1]/2.0+(AOLGBJ[1]+AOLGAJ[1])/2.1+APOCI[1]+APOCJ[1]+(APAH1J[1]+APAH2J[1]+APAH3J[1])/2.03
 AOMIJ ,ug/m3
 ,AXYL1J[1]+AXYL2J[1]+AXYL3J[1]+ATOL1J[1]+ATOL2J[1]+ATOL3J[1]+ABNZ1J[1]+ABNZ2J[1]+ABNZ3J[1]+AISO1J
 [1]+AISO2J[1]+AISO3J[1]+ATRP1J[1]+ATRP2J[1]+ASQTJ[1]+AALK1J[1]+AALK2J[1]+AORGCJ[1]+AOLGBJ[1]+AOLG
 AJ[1]+APOCI[1]+APOCJ[1]+APNCOMI[1]+APNCOMJ[1]+APAH1J[1]+APAH2J[1]+APAH3J[1]
 AORGAJ ,ug/m3
 ,AXYL1J[1]+AXYL2J[1]+AXYL3J[1]+ATOL1J[1]+ATOL2J[1]+ATOL3J[1]+ABNZ1J[1]+ABNZ2J[1]+ABNZ3J[1]+AALK1
 J[1]+AALK2J[1]+AOLGAJ[1]+APAH1J[1]+APAH2J[1]+APAH3J[1]
 AORGBJ ,ug/m3 ,AISO1J[1]+AISO2J[1]+AISO3J[1]+ATRP1J[1]+ATRP2J[1]+ASQTJ[1]+AOLGBJ[1]
 AORGCJ ,ug/m3 ,AORGCJ[1]
 APOCIJ ,ugC/m3 ,APOCI[1]+APOCJ[1]
 APOAIJ ,ug/m3 ,APOCIJ[0]+APNCOMI[1]+APNCOMJ[1]
 ASO4IJ ,ug/m3 ,ASO4I[1]+ASO4J[1]
 ASO4K ,ug/m3 ,ASO4K[1]
 ATOTI ,ug/m3
 ,ASO4I[1]+ANO3I[1]+ANH4I[1]+ANAI[1]+ACLI[1]+AECI[1]+APOCI[1]+APNCOMI[1]+AOTHRI[1]
 ATOTJ ,ug/m3 ,ASO4J[1]+ANO3J[1]+ANH4J[1]+ANAJ[1]+ACLJ[1]+AECJ[1]+AOMIJ[0]-
 (APOCI[1]+APNCOMI[1])+AOTHRJ[1]+AFEJ[1]+ASIJ[1]+ATIJ[1]+ACAJ[1]+AMGJ[1]+AMNJ[1]+AALJ[1]+AKJ[1]
 ATOTK ,ug/m3 ,ASOIL[1]+ACORS[1]+ASEACAT[1]+ACLK[1]+ASO4K[1]+ANO3K[1]+ANH4K[1]
 PMIJ ,ug/m3 ,ATOTI[0]+ATOTJ[0]
 PM10 ,ug/m3 ,PMIJ[0]+ATOTK[0]

```

AUNSPEC1IJ    ,ug/m3    ,PMIJ[0] - (ASOILJ[0] + ANO3IJ[0] + ASO4IJ[0] + ANH4IJ[0] + AOCIJ[0] + AECIJ[0] +
ANAIJ[0] + ACLIJ[0])
ANCOMIJ       ,ug/m3    ,AOMIJ[0]-AOCIJ[0]
AUNSPEC2IJ    ,ug/m3    ,AUNSPEC1IJ[0] - ANCOMIJ[0]
!! OM/OC ratios
AOMOCRAT_PRI  ,none     ,APOAIJ[0]/APOCIJ[0]
AOMOCRAT_TOT  ,none     ,AOMIJ[0]/AOCIJ[0]

```

!! PM2.5 sharp cutoff species

```

PM25_HP       ,ug/m3    ,(AH3OPI[1]*PM25AT[4]+AH3OPJ[1]*PM25AC[4]+AH3OPK[1]*PM25CO[4])*1.0/19.0
PM25_CL       ,ug/m3    ,ACLI[1]*PM25AT[4]+ACLJ[1]*PM25AC[4]+ACLK[1]*PM25CO[4]
PM25_EC       ,ug/m3    ,AECI[1]*PM25AT[4]+AECJ[1]*PM25AC[4]
PM25_NA       ,ug/m3    ,ANAI[1]*PM25AT[4]+ANAJ[1]*PM25AC[4]+ANAK[0]*PM25CO[4]
PM25_MG       ,ug/m3    ,          AMGJ[1]*PM25AC[4]+AMGK[0]*PM25CO[4]
PM25_K        ,ug/m3    ,          AKJ[1]*PM25AC[4]+AKK[0]*PM25CO[4]
PM25_CA       ,ug/m3    ,          ACAJ[1]*PM25AC[4]+ACAK[0]*PM25CO[4]
PM25_NH4      ,ug/m3    ,ANH4I[1]*PM25AT[4]+ANH4J[1]*PM25AC[4]+ANH4K[1]*PM25CO[4]
PM25_NO3      ,ug/m3    ,ANO3I[1]*PM25AT[4]+ANO3J[1]*PM25AC[4]+ANO3K[1]*PM25CO[4]
PM25_OC       ,ugC/m3   ,APOCI[1]*PM25AT[4]+(AOCIJ[0]-APOCI[1])*PM25AC[4]
PM25_SOIL     ,ug/m3    ,ASOILJ[0]*PM25AC[4]+(ASOIL[1]+ACORS[1])*PM25CO[4]
PM25_SO4      ,ug/m3    ,ASO4I[1]*PM25AT[4]+ASO4J[1]*PM25AC[4]+ASO4K[1]*PM25CO[4]
PM25_TOT      ,ug/m3    ,ATOTI[0]*PM25AT[4]+ATOTJ[0]*PM25AC[4]+ATOTK[0]*PM25CO[4]
PM25_UNSPEC1  ,ug/m3    ,PM25_TOT[0]-
(PM25_CL[0]+PM25_EC[0]+PM25_NA[0]+PM25_NH4[0]+PM25_NO3[0]+PM25_OC[0]+PM25_SOIL[0]+PM25_SO4[0])
PMC_CL        ,ug/m3    ,ACLI[1]+ACLJ[1]+ACLK[1]-PM25_CL[0]
PMC_NA        ,ug/m3    ,ANAIJ[0]+ANAK[0]*0.78-PM25_NA[0]
PMC_NH4       ,ug/m3    ,ANH4I[1]+ANH4J[1]+ANH4K[1]-PM25_NH4[0]
PMC_NO3       ,ug/m3    ,ANO3I[1]+ANO3J[1]+ANO3K[1]-PM25_NO3[0]
PMC_SO4       ,ug/m3    ,ASO4I[1]+ASO4J[1]+ASO4K[1]-PM25_SO4[0]
PMC_TOT       ,ug/m3    ,PM10[0]-PM25_TOT[0]

```

!Meteorology

```

DCV_Recon     ,deciview ,DCV_Recon[2]
AIR_DENS      ,kg/m3    ,DENS[3]
RH            ,%        ,100.00*RH[4]
SFC_TMP       ,C        ,(TEMP2[5]-273.15)
PBLH          ,m        ,PBL[5]
SOL_RAD       ,WATTS/m2 ,RGRND[5]
precip        ,cm       ,RN[5]+RC[5]
WSPD10        ,m/s     ,WSPD10[5]
WDIR10        ,deg      ,WDIR10[5]

```

!FRM PM Equivalent Calculation

```

K             ,ppb^2    ,exp(118.87-24084/TEMP2[5]-6.025*log(TEMP2[5]))
P1            ,          ,exp(8763/TEMP2[5]+19.12*log(TEMP2[5])-135.94)
P2            ,          ,exp(9969/TEMP2[5]+16.22*log(TEMP2[5])-122.65)
P3            ,          ,exp(13875/TEMP2[5]+24.46*log(TEMP2[5])-182.61)
a             ,          ,1-RH[0]/100
K_prime       ,ppb^2    ,(P1[0]-P2[0]*a[0]+(P3[0]*a[0]*a[0]))*(a[0]^1.75)*K[0]
sqrt_Ki       ,ppb     ,sqrt(RH[0]<=61 ? K[0] : K_prime[0])
max_NO3_loss  ,ug/m3    ,745.7/TEMP2[5]*sqrt_Ki[0]
PM25_NO3_loss ,ug/m3    ,max_NO3_loss[0]<=PM25_NO3[0] ? max_NO3_loss[0] : PM25_NO3[0]
ANO3IJ_loss   ,ug/m3    ,max_NO3_loss[0]<=ANO3IJ[0] ? max_NO3_loss[0] : ANO3IJ[0]
PM25_NH4_loss ,ug/m3    ,PM25_NO3_loss[0]*(18/62)
ANH4IJ_loss   ,ug/m3    ,ANO3IJ_loss[0]*(18/62)
PMIJ_FRM      ,ug/m3    ,PMIJ[0]-(ANO3IJ_loss[0]+ANH4IJ_loss[0])+0.24*(ASO4IJ[0]+ANH4IJ[0]-ANH4IJ_loss[0])+0.5
PM25_FRM      ,ug/m3    ,PM25_TOT[0]-(PM25_NO3_loss[0]+PM25_NH4_loss[0])+0.24*(PM25_SO4[0]+PM25_NH4[0]-
PM25_NH4_loss[0])+0.5

```

S.3 CMAQv5.1 CB05e51 AE6 Wet/Dry Deposition Species Definitions (available with the CMAQv5.1 release code)

/ File [1]: DRYDEP
 / File [2]: WETDEP
 / File [3]: METCRO2D

/
 /new species ,units ,expression

ANAK_D	,kg/ha	,0.8373*ASEACAT[1]+0.0626*ASOIL[1]+0.0023*ACORS[1]
ANAK_W	,kg/ha	,0.8373*ASEACAT[2]+0.0626*ASOIL[2]+0.0023*ACORS[2]
AMGK_D	,kg/ha	,0.0997*ASEACAT[1] +0.0032*ACORS[1]
AMGK_W	,kg/ha	,0.0997*ASEACAT[2] +0.0032*ACORS[2]
AKK_D	,kg/ha	,0.0310*ASEACAT[1]+0.0242*ASOIL[1]+0.0176*ACORS[1]
AKK_W	,kg/ha	,0.0310*ASEACAT[2]+0.0242*ASOIL[2]+0.0176*ACORS[2]
ACAK_D	,kg/ha	,0.0320*ASEACAT[1]+0.0838*ASOIL[1]+0.0562*ACORS[1]
ACAK_W	,kg/ha	,0.0320*ASEACAT[2]+0.0838*ASOIL[2]+0.0562*ACORS[2]
DDEP_NO2	,kg/ha	,NO2[1]
WDEP_NO2	,kg/ha	,NO2[2]
DDEP_NO	,kg/ha	,NO[1]
WDEP_NO	,kg/ha	,NO[2]
DDEP_FORM	,kg/ha	,FORM[1]
WDEP_FORM	,kg/ha	,FORM[2]
DDEP_H2O2	,kg/ha	,H2O2[1]
WDEP_H2O2	,kg/ha	,H2O2[2]
DDEP_N2O5	,kg/ha	,N2O5[1]
WDEP_N2O5	,kg/ha	,N2O5[2]
DDEP_HONO	,kg/ha	,HONO[1]
WDEP_HONO	,kg/ha	,HONO[2]
DDEP_HNO3	,kg/ha	,HNO3[1]
WDEP_HNO3	,kg/ha	,HNO3[2]
DDEP_ANO3IJ	,kg/ha	,ANO3I[1] + ANO3J[1]
DDEP_ANO3K	,kg/ha	,ANO3K[1]
WDEP_ANO3IJK	,kg/ha	,ANO3I[2] + ANO3J[2] + ANO3K[2]
DDEP_TNO3	,kg/ha	,ANO3I[1] + ANO3J[1] + ANO3K[1] + 0.984*HNO3[1]
WDEP_TNO3	,kg/ha	,ANO3I[2] + ANO3J[2] + ANO3K[2] + 0.984*HNO3[2]
DDEP_NTR	,kg/ha	,NTROH[1]+NTRALK[1]+NTRCN[1]+NTRPX[1]+NTRCNOH[1] + NTRM[1]+NTRI[1]
WDEP_NTR	,kg/ha	,NTROH[2]+NTRALK[2]+NTRCN[2]+NTRPX[2]+NTRCNOH[2] + NTRM[2]+NTRI[2]
DDEP_PANT	,kg/ha	,PAN[1] + PANX[1] + OPAN[1] + MAPAN[1]
WDEP_PANT	,kg/ha	,PAN[2] + PANX[2] + OPAN[2] + MAPAN[2]
DDEP_NH3	,kg/ha	,NH3[1]
WDEP_NH3	,kg/ha	,NH3[2]
DDEP_ANH4IJ	,kg/ha	,ANH4I[1] + ANH4J[1]
DDEP_ANH4K	,kg/ha	,ANH4K[1]
WDEP_ANH4IJK	,kg/ha	,ANH4I[2] + ANH4J[2] + ANH4K[2]
DDEP_NHX	,kg/ha	,ANH4I[1] + ANH4J[1] + ANH4K[1] + 1.059*NH3[1]
WDEP_NHX	,kg/ha	,ANH4I[2] + ANH4J[2] + ANH4K[2] + 1.059*NH3[2]
DDEP_SO2	,kg/ha	,SO2[1]
DDEP_ASO4IJ	,kg/ha	,ASO4I[1] + ASO4J[1]
DDEP_ASO4K	,kg/ha	,ASO4K[1]
WDEP_ASO4IJK	,kg/ha	,ASO4I[2] + ASO4J[2] + ASO4K[2]
WDEP_TSO4	,kg/ha	,ASO4I[2] + ASO4J[2] + ASO4K[2] + 1.5*SO2[2]
DDEP_AECIJ	,kg/ha	,AECI[1] + AECJ[1]
DDEP_AOCIJ	,kg/ha	
,(AXYL1J[1]+AXYL2J[1]+AXYL3J[1])/2.0+(ATOL1J[1]+ATOL2J[1]+ATOL3J[1])/2.0+(ABNZ1J[1]+ABNZ2J[1]+ABNZ3J[1])/2.0+(AISO1J[1]+AISO2J[1])/1.6+AISO3J[1]/2.7+(ATRP1J[1]+ATRP2J[1])/1.4+ASQTJ[1]/2.1+0.64*(AALK1J[1]+AALK2J[1])+(APAH1J[1]+APAH2J[1]+APAH3J[1])/2.03+AORGCJ[1]/2.0 +(AOLGBJ[1]+AOLGAJ[1])/2.1+APOCI[1]+APOCJ[1]		
DDEP_SSSO4J	,kg/ha	,0.19579876*ANAJ[1]

DDEP_SSSO4K ,kg/ha ,0.19579876*ANAK_D[0]
WDEP_SSSO4JK ,kg/ha ,0.19579876*ANAJ[2] + 0.19579876*ANAK_W[0]
DDEP_ANAJ ,kg/ha ,ANAJ[1]
DDEP_ANAK ,kg/ha ,ANAK_D[0]
DDEP_ANAJK ,kg/ha ,ANAJ[1] + ANAK_D[0]
WDEP_ANAJK ,kg/ha ,ANAJ[2] + ANAK_W[0]
TDEP_ANAJK ,kg/ha ,DDEP_ANAJK[0] + WDEP_ANAJK[0]
DDEP_ACLJ ,kg/ha ,ACLJ[1]
DDEP_ACLK ,kg/ha ,ACLK[1]
DDEP_ACLJK ,kg/ha ,ACLJ[1] + ACLK[1]
WDEP_TCL ,kg/ha ,0.972*HCL[2]+0.435*CLNO2[2] + ACLJ[2] + ACLK[2]
TDEP_CL ,kg/ha ,DDEP_ACLJK[0] + WDEP_TCL[0]
DDEP_CAJ ,kg/ha ,ACAJ[1]
WDEP_CAJ ,kg/ha ,ACAJ[2]
DDEP_CAJK ,kg/ha ,ACAJ[1]+ACAK_D[0]
WDEP_CAJK ,kg/ha ,ACAJ[2]+ACAK_W[0]
DDEP_FEJ ,kg/ha ,AFEJ[1]
WDEP_FEJ ,kg/ha ,AFEJ[2]
DDEP_ALJ ,kg/ha ,AALJ[1]
WDEP_ALJ ,kg/ha ,AALJ[2]
DDEP_SIJ ,kg/ha ,ASIJ[1]
WDEP_SIJ ,kg/ha ,ASIJ[2]
DDEP_TIJ ,kg/ha ,ATIJ[1]
WDEP_TIJ ,kg/ha ,ATIJ[2]
DDEP_MGJ ,kg/ha ,AMGJ[1]
WDEP_MGJ ,kg/ha ,AMGJ[2]
DDEP_MGJK ,kg/ha ,AMGJ[1]+AMGK_D[0]
WDEP_MGJK ,kg/ha ,AMGJ[2]+AMGK_W[0]
DDEP_KJ ,kg/ha ,AKJ[1]
WDEP_KJ ,kg/ha ,AKJ[2]
DDEP_KJK ,kg/ha ,AKJ[1]+AKK_D[0]
WDEP_KJK ,kg/ha ,AKJ[2]+AKK_W[0]
DDEP_MNJ ,kg/ha ,AMNJ[1]
WDEP_MNJ ,kg/ha ,AMNJ[2]
DDEP_O3 ,kg/ha ,O3[1]
WDEP_O3 ,kg/ha ,O3[2]
WDEP_PNA ,kg/ha ,PNA[2]
RT ,cm ,RN[3] + RC[3]
DD_OXN_NOX ,kg/ha ,0.30435*NO2[1] + 0.46667*NO[1]
WD_OXN_NOX ,kg/ha ,0.30435*NO2[2] + 0.46667*NO[2]
DD_OXN_TNO3 ,kg/ha ,0.22222*HNO3[1] + 0.22581*ANO3I[1] + 0.22581*ANO3J[1] + 0.22581*ANO3K[1]
WD_OXN_TNO3 ,kg/ha ,0.22581*WDEP_TNO3[0]
DD_OXN_PANT ,kg/ha ,0.11570*PAN[1] + 0.11570*PANX[1] + 0.11570*OPAN[1] + 0.11570*MAPAN[1]
WD_OXN_PANT ,kg/ha ,0.11570*PAN[2] + 0.11570*PANX[2] + 0.11570*OPAN[2] + 0.11570*MAPAN[2]
DD_OXN_ORGN ,kg/ha ,0.10770*(NTROH[1]+NTRALK[1]+NTRCN[1]+NTRPX[1]+NTRCNOH[1] +
NTRM[1]+NTRI[1])
WD_OXN_ORGN ,kg/ha ,0.10770*(NTROH[2]+NTRALK[2]+NTRCN[2]+NTRPX[2]+NTRCNOH[2] +
NTRM[2]+NTRI[2])
/DD_OXN_OTHR ,kg/ha ,0.25926*N2O5[1] + 0.29787*HONO[1]+0.1717*CLNO2[1]
WD_OXN_OTHR ,kg/ha ,0.25926*N2O5[2] + 0.29787*HONO[2]+0.177720*PNA[2]+0.1717*CLNO2[2]
/DD_OXN_TOT ,kg/ha ,DD_OXN_NOX[0] + DD_OXN_TNO3[0] + DD_OXN_PANT[0] + DD_OXN_ORGN[0] +
DD_OXN_OTHR[0]
WD_OXN_TOT ,kg/ha ,WD_OXN_NOX[0] + WD_OXN_TNO3[0] + WD_OXN_PANT[0] + WD_OXN_ORGN[0] +
WD_OXN_OTHR[0]
/TD_OXN_TOT ,kg/ha ,DD_OXN_TOT[0] + WD_OXN_TOT[0]
/DD_OXN_TOTMEQ ,meq/m2 ,7.14*DD_OXN_TOT[0]
WD_OXN_TOTMEQ ,meq/m2 ,7.14*WD_OXN_TOT[0]
/TD_OXN_TOTMEQ ,meq/m2 ,DD_OXN_TOTMEQ[0] + WD_OXN_TOTMEQ[0]
DD_REDN_TOT ,kg/ha ,0.7777*DDEP_NHX[0]
WD_REDN_TOT ,kg/ha ,0.7777*WDEP_NHX[0]

TD_REDN_TOT ,kg/ha ,DD_REDN_TOT[0] + WD_REDN_TOT[0]
 DD_REDN_TOTMEQ ,meq/m2 ,7.14*DD_REDN_TOT[0]
 WD_REDN_TOTMEQ ,meq/m2 ,7.14*WD_REDN_TOT[0]
 TD_REDN_TOTMEQ ,meq/m2 ,DD_REDN_TOTMEQ[0] + WD_REDN_TOTMEQ[0]
 DD_S_TOT ,kg/ha ,0.5*SO2[1] + 0.33333*ASO4I[1] + 0.33333*ASO4J[1] + 0.33333*ASO4K[1]
 WD_S_TOT ,kg/ha ,0.33333*WDEP_TSO4[0]
 TD_S_TOT ,kg/ha ,DD_S_TOT[0] + WD_S_TOT[0]
 DD_S_TOTMEQ ,meq/m2 ,6.24*DD_S_TOT[0]
 WD_S_TOTMEQ ,meq/m2 ,6.24*WD_S_TOT[0]
 TD_S_TOTMEQ ,meq/m2 ,DD_S_TOTMEQ[0] + WD_S_TOTMEQ[0]
 DD_S_SeaS ,kg/ha ,0.33333*DDEP_SSSO4J[0] + 0.33333*DDEP_SSSO4K[0]
 WD_S_SeaS ,kg/ha ,0.33333*WDEP_SSSO4JK[0]
 TD_S_SeaS ,kg/ha ,DD_S_SeaS[0] + WD_S_SeaS[0]
 DD_S_SeaSMEQ ,meq/m2 ,6.24*DD_S_SeaS[0]
 WD_S_SeaSMEQ ,meq/m2 ,6.24*WD_S_SeaS[0]
 TD_S_SeaSMEQ ,meq/m2 ,DD_S_SeaSMEQ[0] + WD_S_SeaSMEQ[0]

S.4 – WRFv3.4 Namelist

```
&time_control
start_year      = 2011,
start_month     = 12,
start_day       = 27,
start_hour      = 00,
start_minute    = 00,
start_second    = 00,
end_year        = 2012,
end_month       = 01,
end_day         = 01,
end_hour        = 00,
end_minute      = 00,
end_second      = 00,
interval_seconds = 10800,
input_from_file = .true.,
history_interval = 60,
frames_per_outfile = 24,
restart         = .TRUE.,
restart_interval = 7200,
io_form_history = 2
io_form_restart = 2
io_form_input   = 2
io_form_boundary = 2
debug_level     = 0
io_form_auxinput2 = 2
io_form_auxinput4 = 2
auxinput1_inname = "metoa_em.d01.<date>"
auxinput4_inname = "wrflowinp_d01"
auxinput4_interval = 180
auxinput4_end_h   = 9001
write_hist_at_0h_rst = .true.,
/

&domains
time_step      = 60,
time_step_fract_num = 0,
time_step_fract_den = 1,
use_adaptive_time_step = .false.
max_dom        = 1,
s_we           = 1,
e_we           = 472,
s_sn           = 1,
e_sn           = 312,
s_vert         = 1,
e_vert         = 36,
p_top_requested = 5000,
eta_levels     = 1.000, 0.9975, 0.995, 0.990, 0.985,
               0.980, 0.970, 0.960, 0.950,
               0.940, 0.930, 0.920, 0.910,
               0.900, 0.880, 0.860, 0.840,
               0.820, 0.800, 0.770, 0.740,
               0.700, 0.650, 0.600, 0.550,
               0.500, 0.450, 0.400, 0.350,
               0.300, 0.250, 0.200, 0.150,
               0.100, 0.050, 0.000
dx             = 12000,
dy             = 12000,
grid_id        = 1,
```

```

parent_id           = 0,
i_parent_start      = 0,
j_parent_start      = 0,
parent_grid_ratio    = 1,
parent_time_step_ratio = 1,
feedback            = 1,
smooth_option       = 0,
/

```

```

&physics
mp_physics          = 10,
ra_lw_physics       = 4,
ra_sw_physics       = 4,
radt               = 20,
sf_sfclay_physics   = 7,
sf_surface_physics  = 7,
bl_pbl_physics      = 7,
bldt               = 0,
cu_physics          = 1,
kfeta_trigger       = 2,
cudt               = 0,
isfflx             = 1,
ifsnow             = 1,
icloud             = 1,
surface_input_source = 1,
num_soil_layers     = 2,
sst_update         = 1,
pxlsm_smois_init    = 0,
slope_rad          = 1,
topo_shading        = 1,
shadlen            = 25000.,
num_land_cat        = 40,
prec_acc_dt         = 60,
mp_zero_out         = 2,
fractional_seaice    = 1,
seaice_threshold    = 0.0,
/

```

```

&fdda
grid_fdda           = 1,
grid_sfdda          = 1,
pxlsm_soil_nudge    = 1,
sgfdda_inname       = "wrfsfdda_d01",
sgfdda_end_h        = 9001,
sgfdda_interval_m   = 180,
sgfdda_interval     = 10800,
gfdda_inname        = "wrffdda_d<domain>",
gfdda_end_h         = 9001,
gfdda_interval_m    = 180,
fgdt                = 0,
if_no_pbl_nudging_uv = 1,
if_no_pbl_nudging_t = 1,
if_no_pbl_nudging_q = 1,
if_zfac_uv          = 0,
k_zfac_uv           = 13,
if_zfac_t           = 0,
k_zfac_t            = 13,
if_zfac_q           = 0,
k_zfac_q            = 13,
guv                 = 0.0001,

```



```

gt                = 0.0001,
gq                = 0.00001,
guv_sfc           = 0.0000,
gt_sfc            = 0.0000,
gq_sfc            = 0.0000,
if_ramping        = 0,
dtramp_min        = 60.0,
io_form_gfdda     = 2,
rinblw            = 250.0
/

```

```

&dynamics
w_damping         = 1,
diff_opt          = 1,
km_opt            = 4,
diff_6th_opt      = 2,
diff_6th_factor   = 0.12,
damp_opt          = 3,
base_temp         = 290.
zdamp             = 5000.,
dampcoef          = 0.05,
khdif             = 0,
kvdif             = 0,
non_hydrostatic   = .true.,
moist_adv_opt     = 2,
tke_adv_opt       = 2,
scalar_adv_opt    = 2,
/

```

```

&dfi_control
dfi_opt           = 0
dfi_nfilter       = 7
dfi_write_filtered_input = .true.
dfi_write_dfi_history = .false.
dfi_cutoff_seconds = 60
dfi_time_dim      = 1000
dfi_bckstop_year  = 2006
dfi_bckstop_month = 08
dfi_bckstop_day   = 04
dfi_bckstop_hour  = 12
dfi_bckstop_minute = 00
dfi_bckstop_second = 00
dfi_fwdstop_year  = 2006
dfi_fwdstop_month = 08
dfi_fwdstop_day   = 04
dfi_fwdstop_hour  = 13
dfi_fwdstop_minute = 00
dfi_fwdstop_second = 00
/

```

```

&bdy_control
spec_bdy_width    = 5,
spec_zone         = 1,
relax_zone        = 4,
specified         = .true.,
nested            = .false.,
/

```

```

&grib2
/

```

```
&namelist_quilt  
nio_tasks_per_group = 0,  
nio_groups = 1,  
/
```

S.5 – WRFv3.7 Namelist

```
&time_control
start_year      = 2011,
start_month     = 12,
start_day       = 27,
start_hour      = 00,
start_minute    = 00,
start_second    = 00,
end_year        = 2012,
end_month       = 01,
end_day         = 01,
end_hour        = 00,
end_minute      = 00,
end_second      = 00,
interval_seconds = 10800,
input_from_file = .true.,
history_interval = 60,
frames_per_outfile = 24,
restart         = .TRUE.,
restart_interval = 7200,
io_form_history = 2
io_form_restart = 2
io_form_input   = 2
io_form_boundary = 2
debug_level     = 0
io_form_auxinput2 = 2
io_form_auxinput4 = 2
auxinput1_inname = "metoa_em.d01.<date>"
auxinput4_inname = "wrflowinp_d01"
auxinput4_interval = 180
auxinput4_end_h   = 9001
write_hist_at_0h_rst = .true.,
/

&domains
time_step      = 60,
time_step_fract_num = 0,
time_step_fract_den = 1,
use_adaptive_time_step = .false.
max_dom        = 1,
s_we           = 1,
e_we           = 472,
s_sn           = 1,
e_sn           = 312,
s_vert         = 1,
e_vert         = 36,
p_top_requested = 5000,
eta_levels     = 1.000, 0.9975, 0.995, 0.990, 0.985,
               0.980, 0.970, 0.960, 0.950,
               0.940, 0.930, 0.920, 0.910,
               0.900, 0.880, 0.860, 0.840,
               0.820, 0.800, 0.770, 0.740,
               0.700, 0.650, 0.600, 0.550,
               0.500, 0.450, 0.400, 0.350,
               0.300, 0.250, 0.200, 0.150,
               0.100, 0.050, 0.000
dx             = 12000,
dy             = 12000,
grid_id        = 1,
```

```

parent_id          = 0,
i_parent_start     = 0,
j_parent_start     = 0,
parent_grid_ratio  = 1,
parent_time_step_ratio = 1,
feedback           = 1,
smooth_option      = 0,
/

```

```

&physics
mp_physics          = 10,
ra_lw_physics       = 4,
ra_sw_physics       = 4,
radt                = 20,
sf_sfclay_physics   = 7,
sf_surface_physics  = 7,
bl_pbl_physics      = 7,
bldt                = 0,
cu_physics          = 1,
kfeta_trigger       = 2,
cudt                = 0,
isfflx              = 1,
ifsnow              = 1,
icloud              = 1,
cu_rad_feedback     = .true.,
surface_input_source = 1,
num_soil_layers     = 2,
sst_update          = 1,
pxlsm_smois_init    = 0,
slope_rad           = 1,
topo_shading        = 1,
shadlen             = 25000.,
num_land_cat        = 40,
prec_acc_dt         = 60,
mp_zero_out         = 2,
fractional_seaice    = 1,
seaice_threshold    = 0.0,
/

```

```

&fdda
grid_fdda           = 1,
grid_sfdda          = 1,
pxlsm_soil_nudge    = 1,
sgfdda_inname       = "wrfsfdda_d01",
sgfdda_end_h        = 9001,
sgfdda_interval_m   = 180,
sgfdda_interval     = 10800,
gfdda_inname        = "wrffdda_d<domain>",
gfdda_end_h         = 9001,
gfdda_interval_m    = 180,
fgdt                 = 0,
if_no_pbl_nudging_uv = 1,
if_no_pbl_nudging_t = 1,
if_no_pbl_nudging_q = 1,
if_zfac_uv          = 0,
k_zfac_uv           = 13,
if_zfac_t           = 0,
k_zfac_t            = 13,
if_zfac_q           = 0,
k_zfac_q            = 13,

```

```

guv                = 0.0001,
gt                 = 0.0001,
gq                 = 0.00001,
guv_sfc            = 0.0000,
gt_sfc             = 0.0000,
gq_sfc             = 0.0000,
if_ramping         = 0,
dtramp_min         = 60.0,
io_form_gfdda      = 2,
rinblw             = 250.0
/

```

```

&dynamics
w_damping          = 1,
diff_opt           = 1,
km_opt             = 4,
diff_6th_opt       = 2,
diff_6th_factor    = 0.12,
damp_opt           = 3,
base_temp          = 290.
zdamp              = 5000.,
dampcoef           = 0.05,
khdif              = 0,
kvdif              = 0,
non_hydrostatic    = .true.,
moist_adv_opt      = 2,
tke_adv_opt        = 2,
scalar_adv_opt     = 2,
/

```

```

&dfi_control
dfi_opt            = 0
dfi_nfilter        = 7
dfi_write_filtered_input = .true.
dfi_write_dfi_history = .false.
dfi_cutoff_seconds = 60
dfi_time_dim       = 1000
dfi_bckstop_year   = 2006
dfi_bckstop_month  = 08
dfi_bckstop_day    = 04
dfi_bckstop_hour   = 12
dfi_bckstop_minute = 00
dfi_bckstop_second = 00
dfi_fwdstop_year   = 2006
dfi_fwdstop_month  = 08
dfi_fwdstop_day    = 04
dfi_fwdstop_hour   = 13
dfi_fwdstop_minute = 00
dfi_fwdstop_second = 00
/

```

```

&bdy_control
spec_bdy_width     = 5,
spec_zone          = 1,
relax_zone         = 4,
specified          = .true.,
nested             = .false.,
/

```

```

&grib2

```

/

```
&namelist_quilt  
nio_tasks_per_group = 0,  
nio_groups = 1,  
/
```

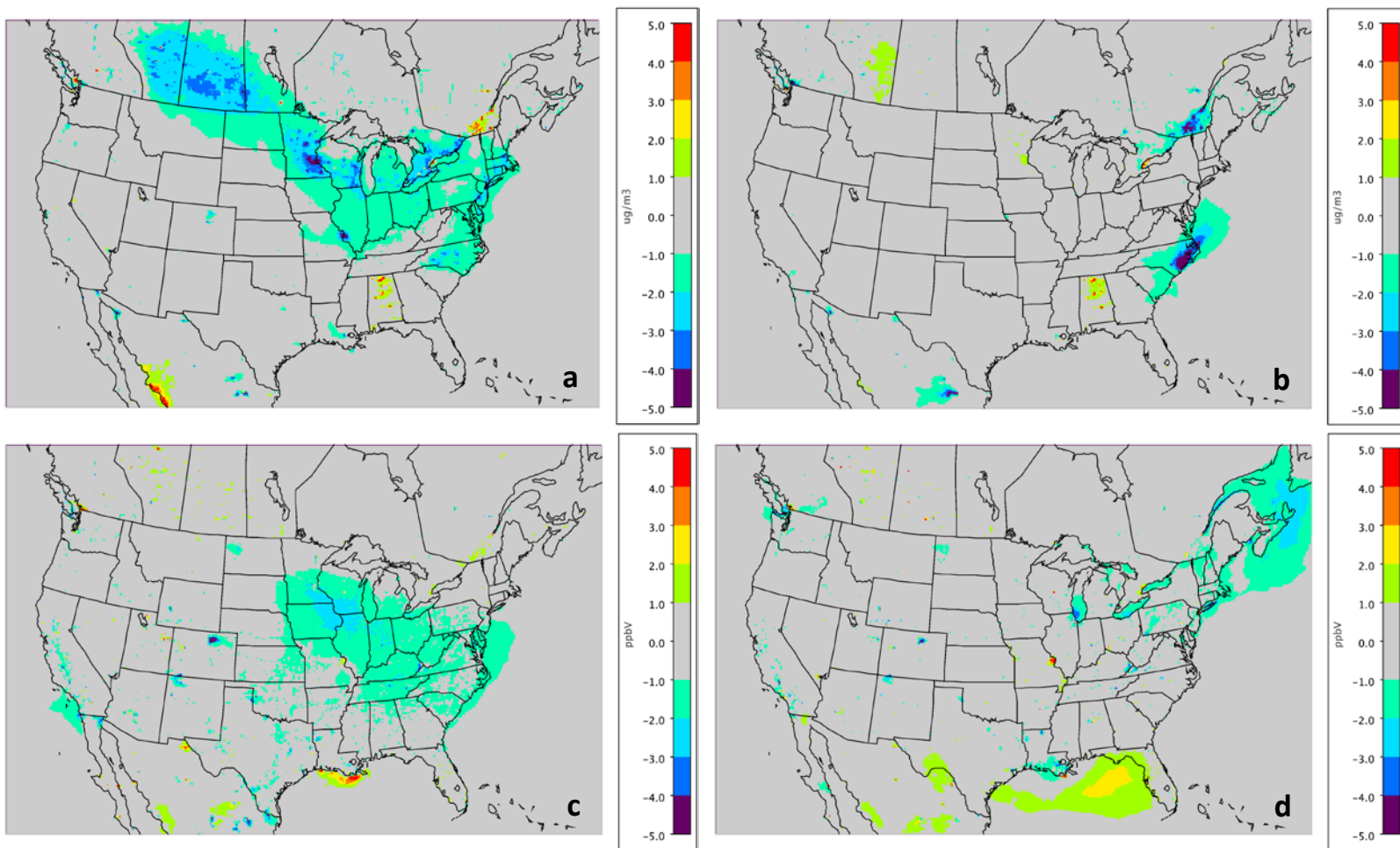


Figure S1: Difference in monthly average PM_{2.5} (ugm⁻³) for January (a) and July (b) and O₃ (ppbV) for January (c) and July (d) between the version 1 and version 2 emissions platform used.

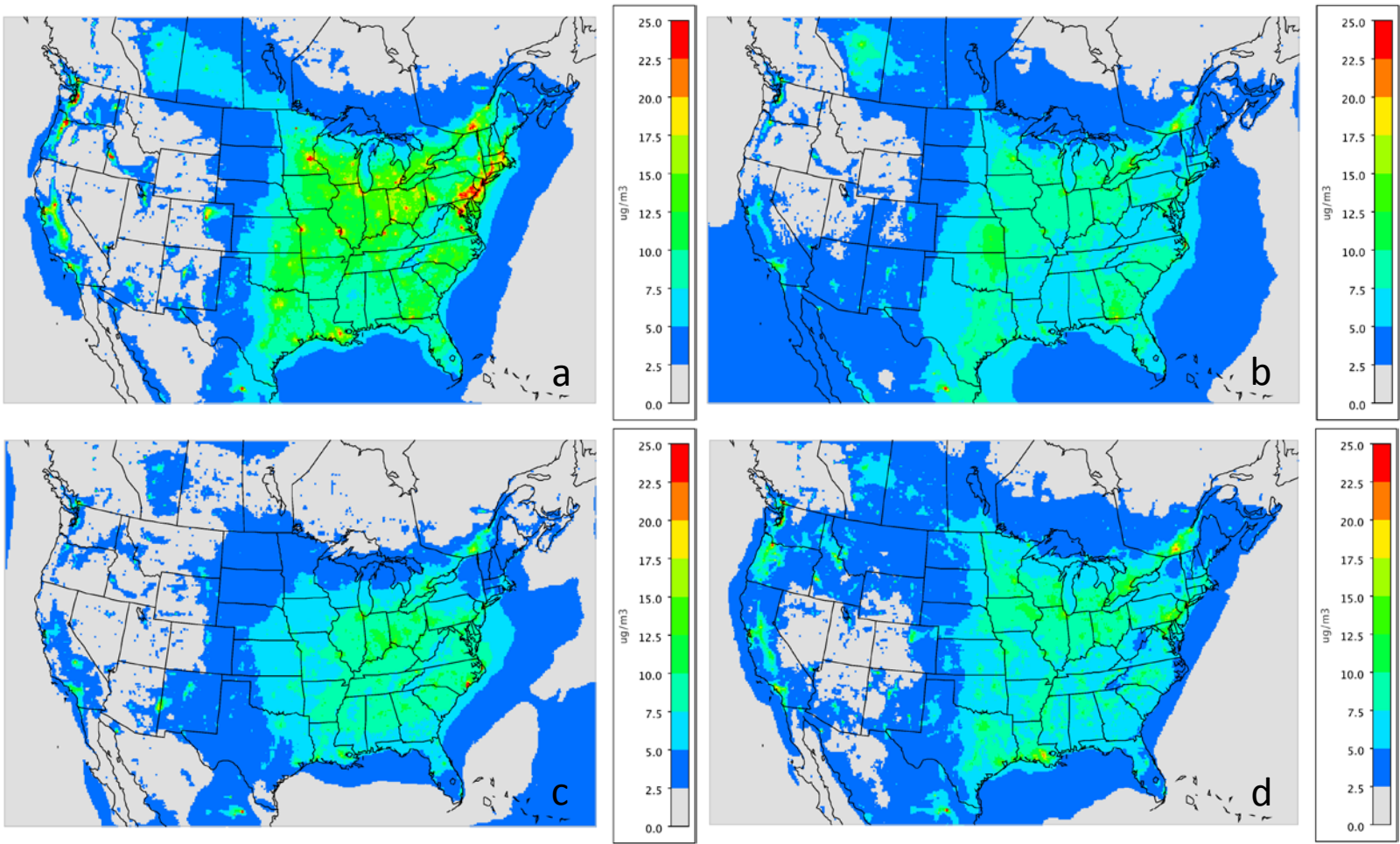


Figure S2: CMAQv5.0.2 simulation seasonal average PM_{2.5} concentrations ($\mu\text{g}/\text{m}^3$) for a) winter b) spring c) summer and d) fall.

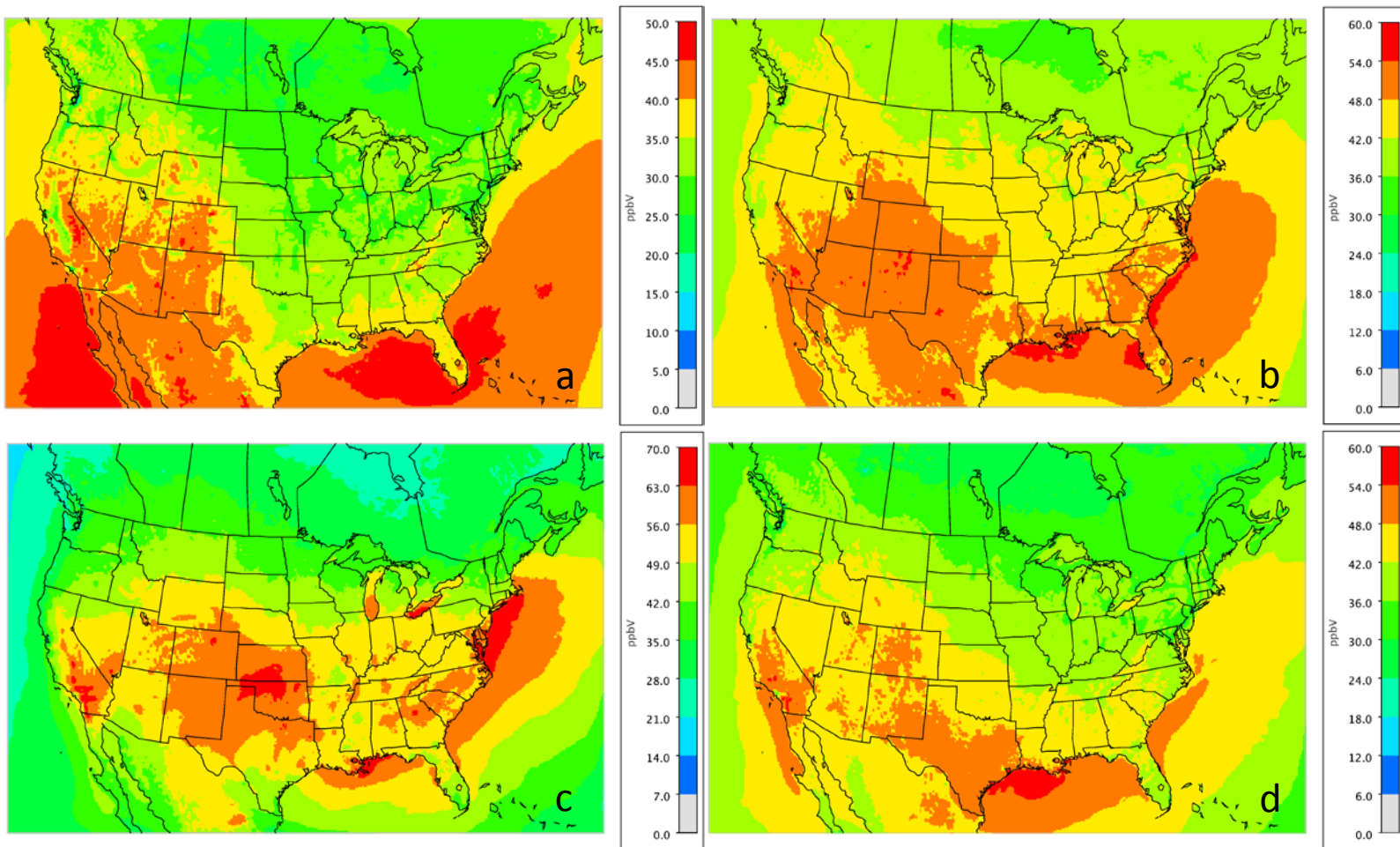


Figure S3: CMAQv5.0.2 simulation seasonal average MDA8 O₃ mixing ratio (ppbv) for a) winter b) spring c) summer and d) fall. Note that the scales for each plot can vary.

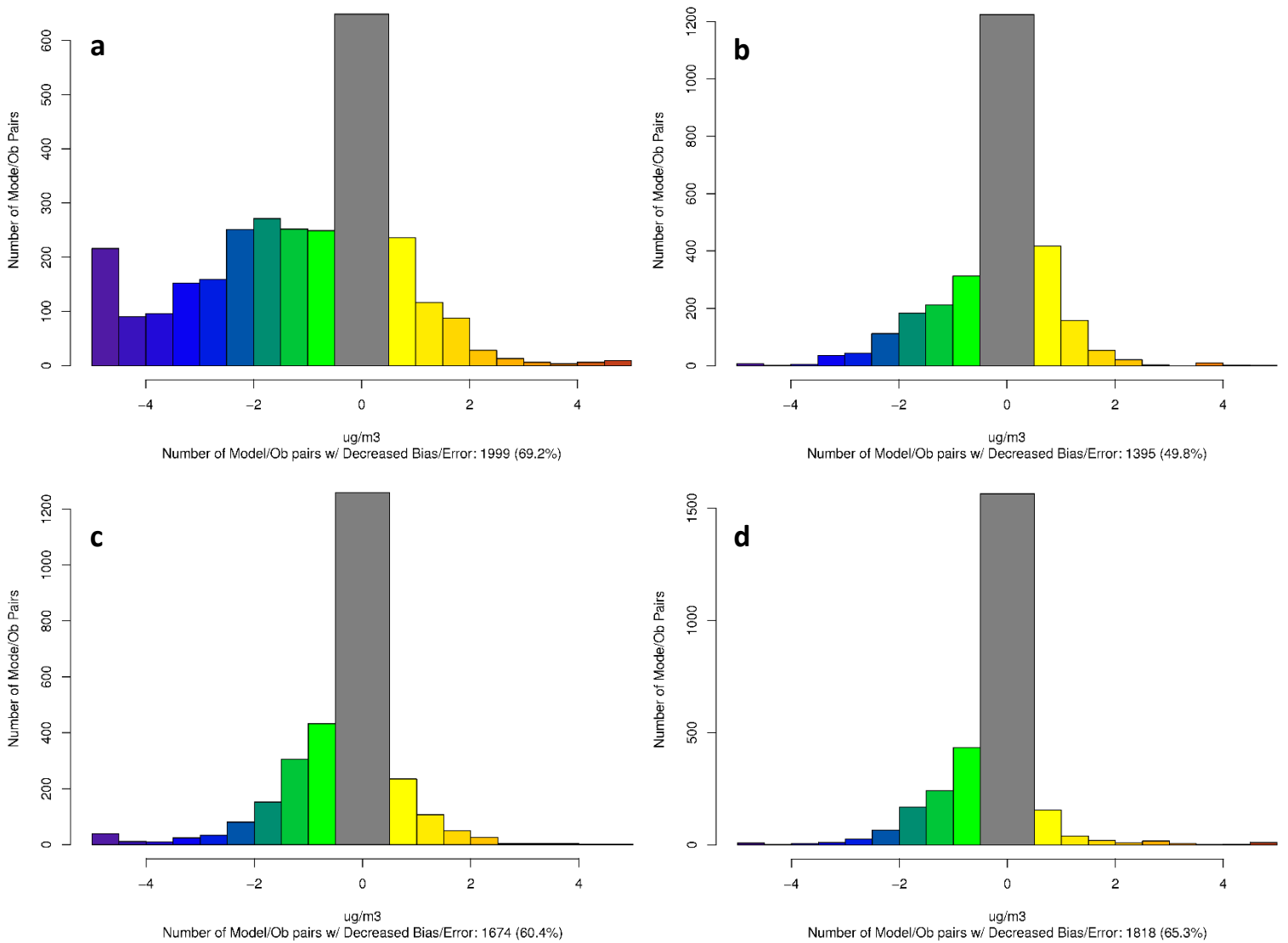


Figure S4: Histograms of the difference in the absolute value of monthly average (2011) PM_{2.5} mean bias for winter (DJF; top left), spring (MAM; top right), summer (JJA; bottom left) and fall (SON; bottom right) between CMAQ v5.0.2_Base and v5.1_Base (CMAQv5.1_Base – CMAQv5.0.2_Base). All plots are in units of $\mu\text{g}/\text{m}^3$. Cool colors indicate a reduction in PM_{2.5} mean bias in CMAQv5.1_Base while warm color indicate an increase in PM_{2.5} mean bias CMAQv5.1_Base.

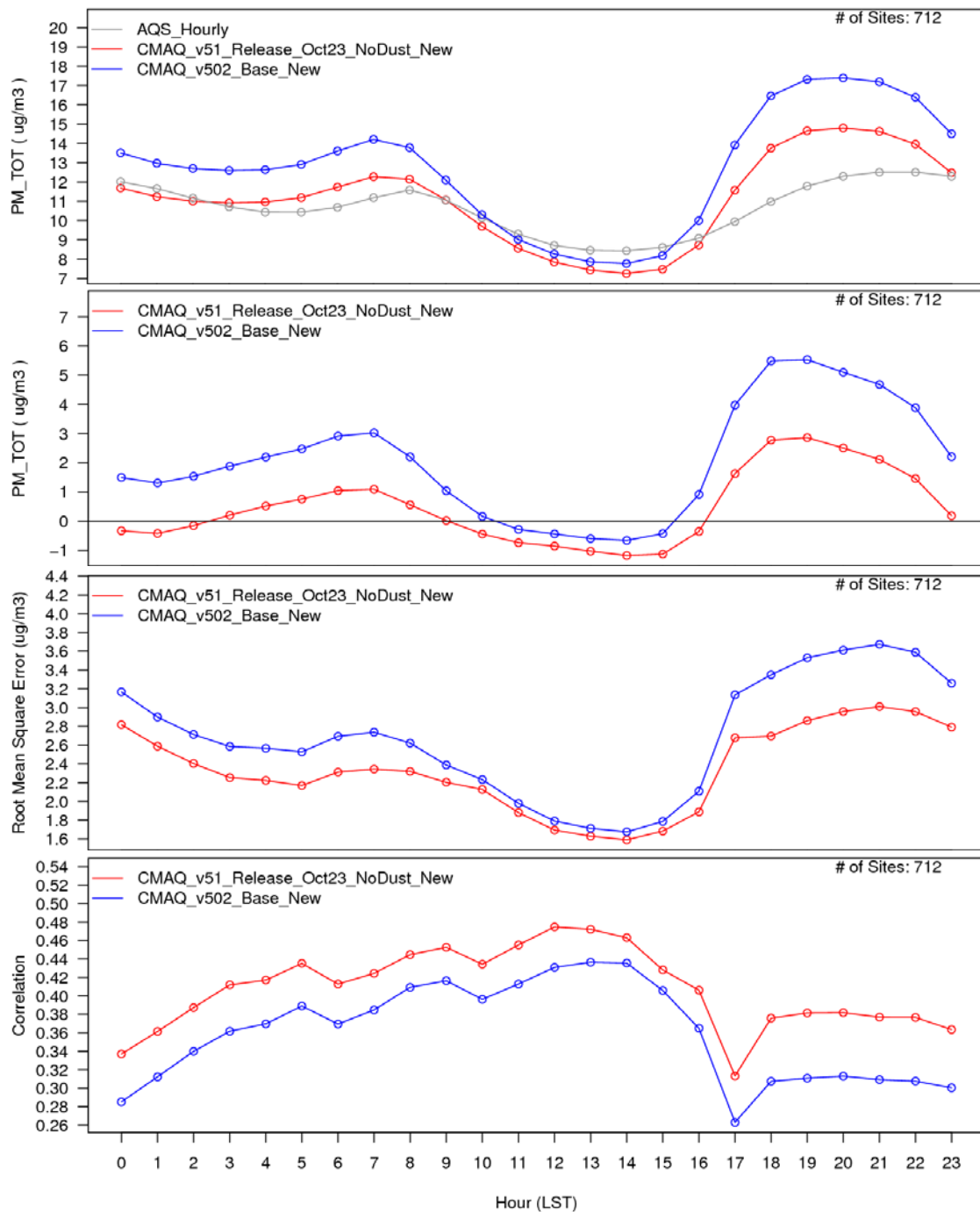


Figure S5: Diurnal time series of winter PM_{2.5} from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in µg m⁻³ except for correlation.

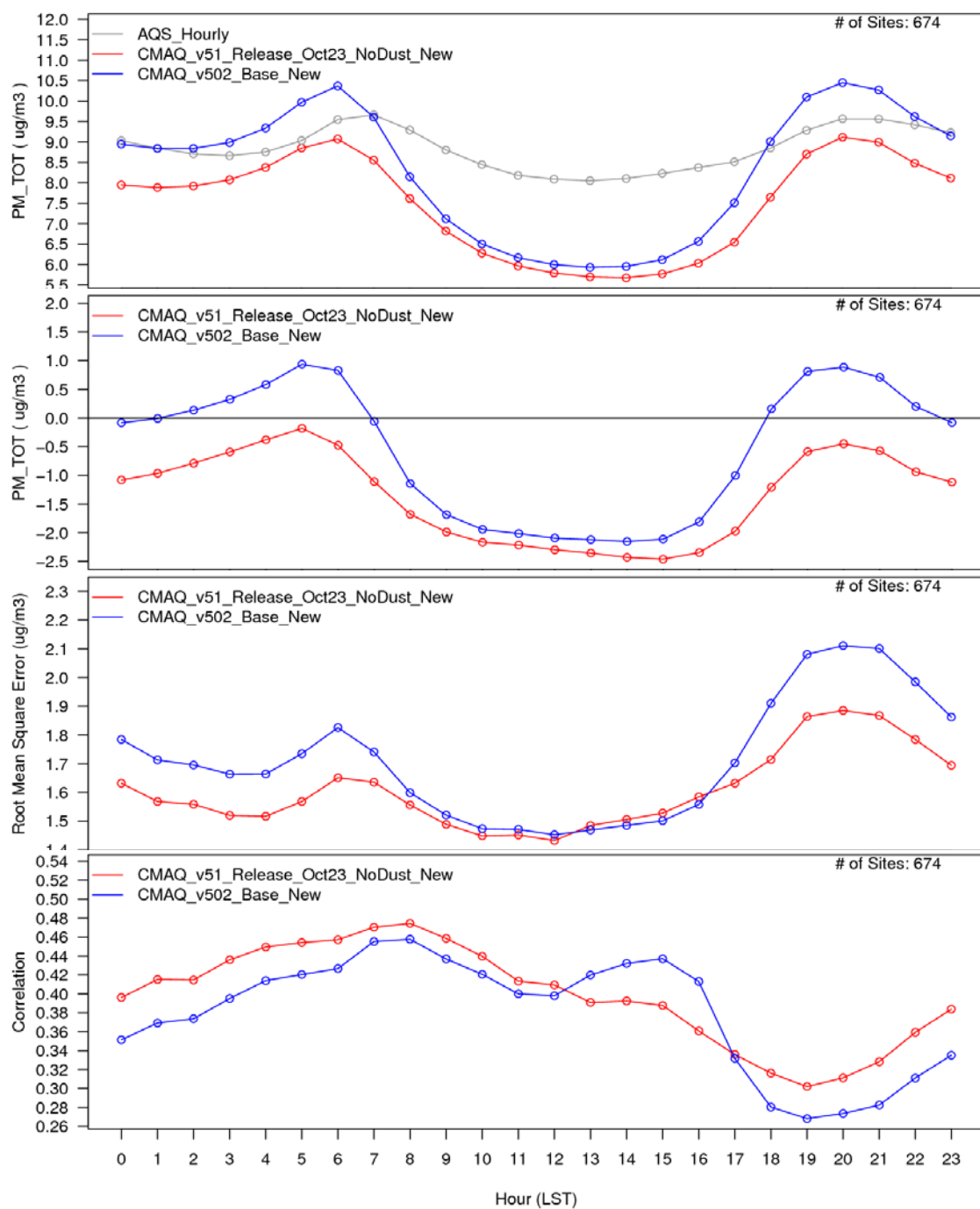


Figure S6: Diurnal time series of spring PM_{2.5} from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in $\mu\text{g m}^{-3}$ except for correlation.

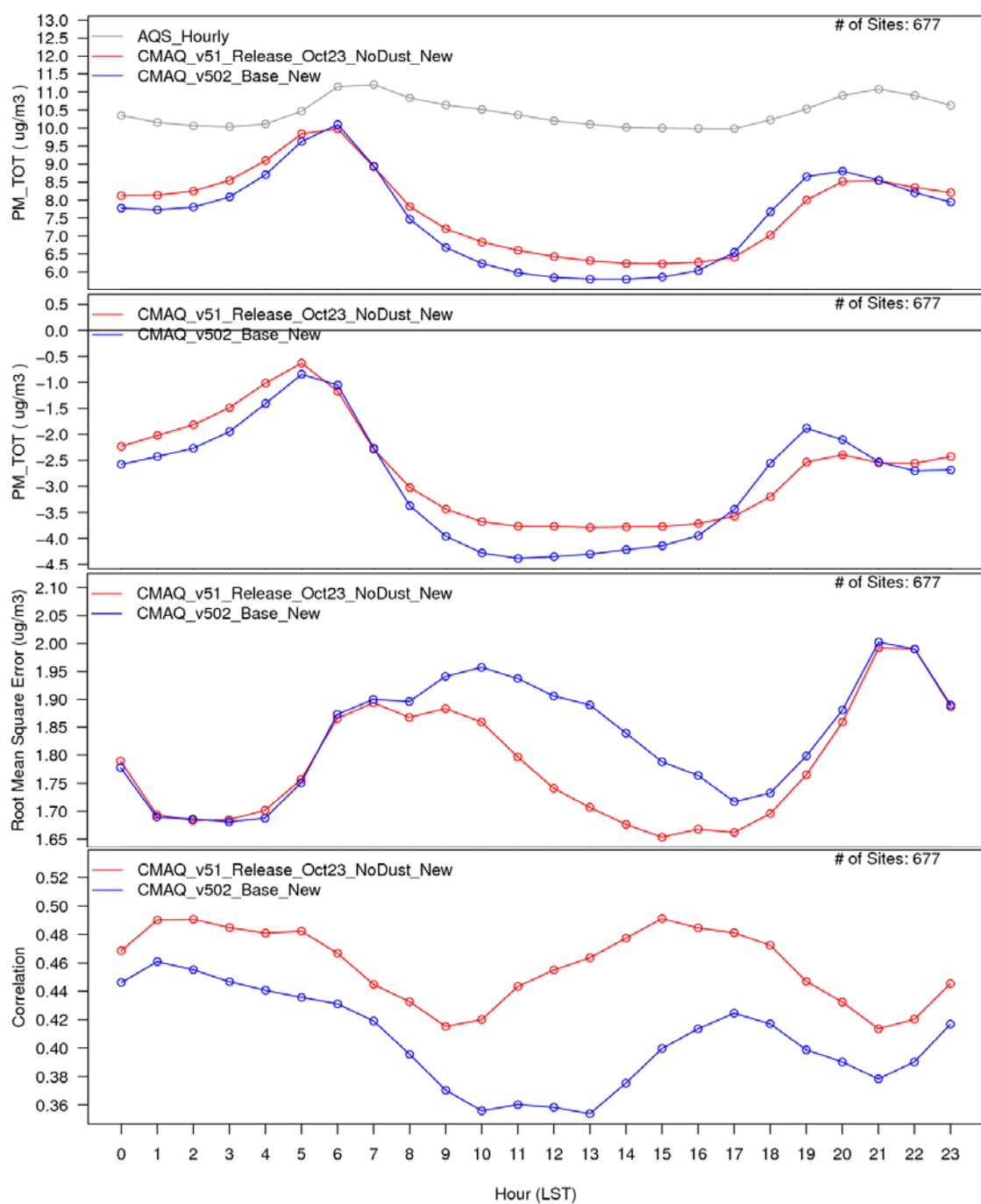


Figure S7: Diurnal time series of summer PM_{2.5} from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in $\mu\text{g m}^{-3}$ except for correlation.

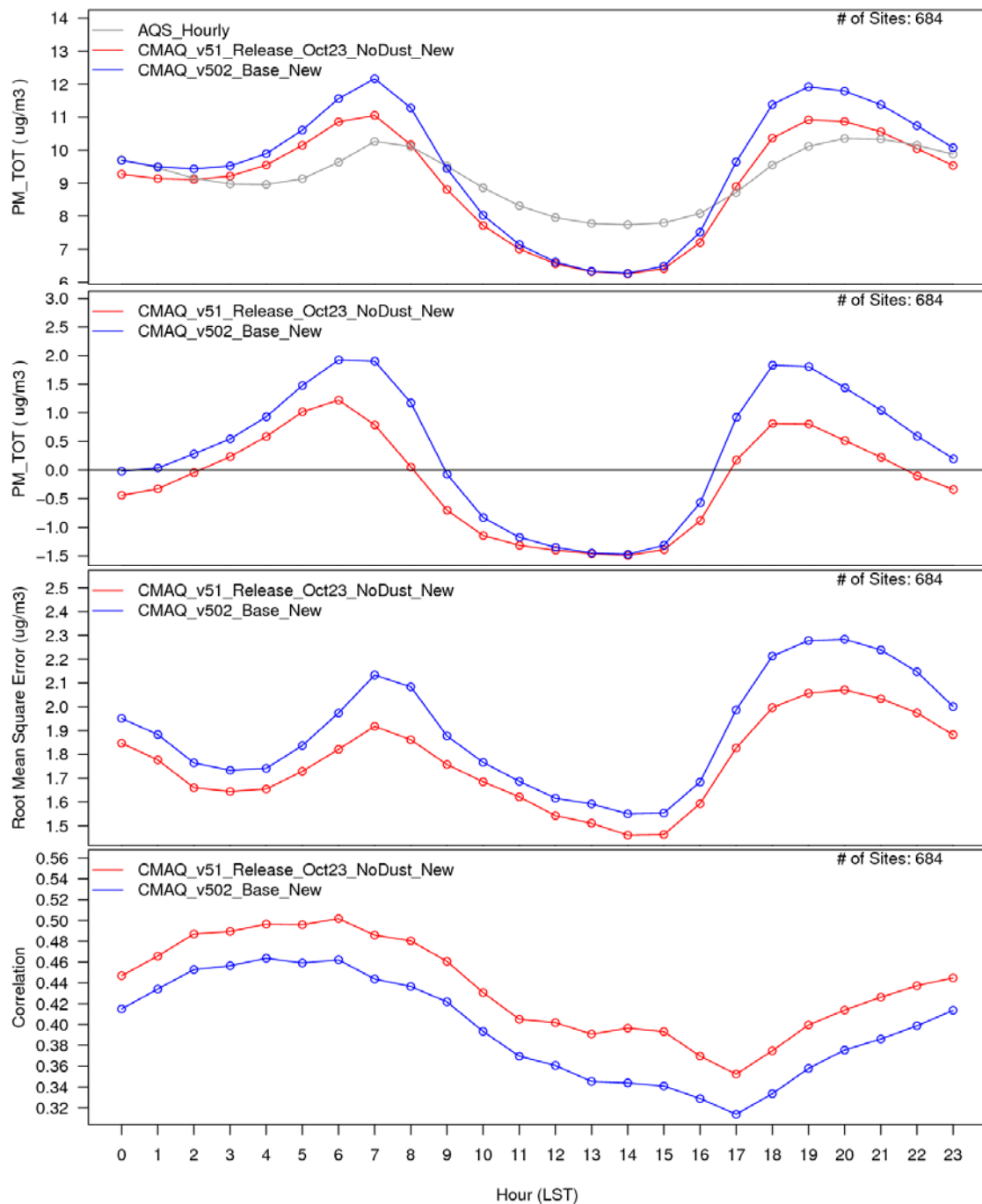


Figure S8: Diurnal time series of fall PM_{2.5} from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in µgm⁻³ except for correlation.

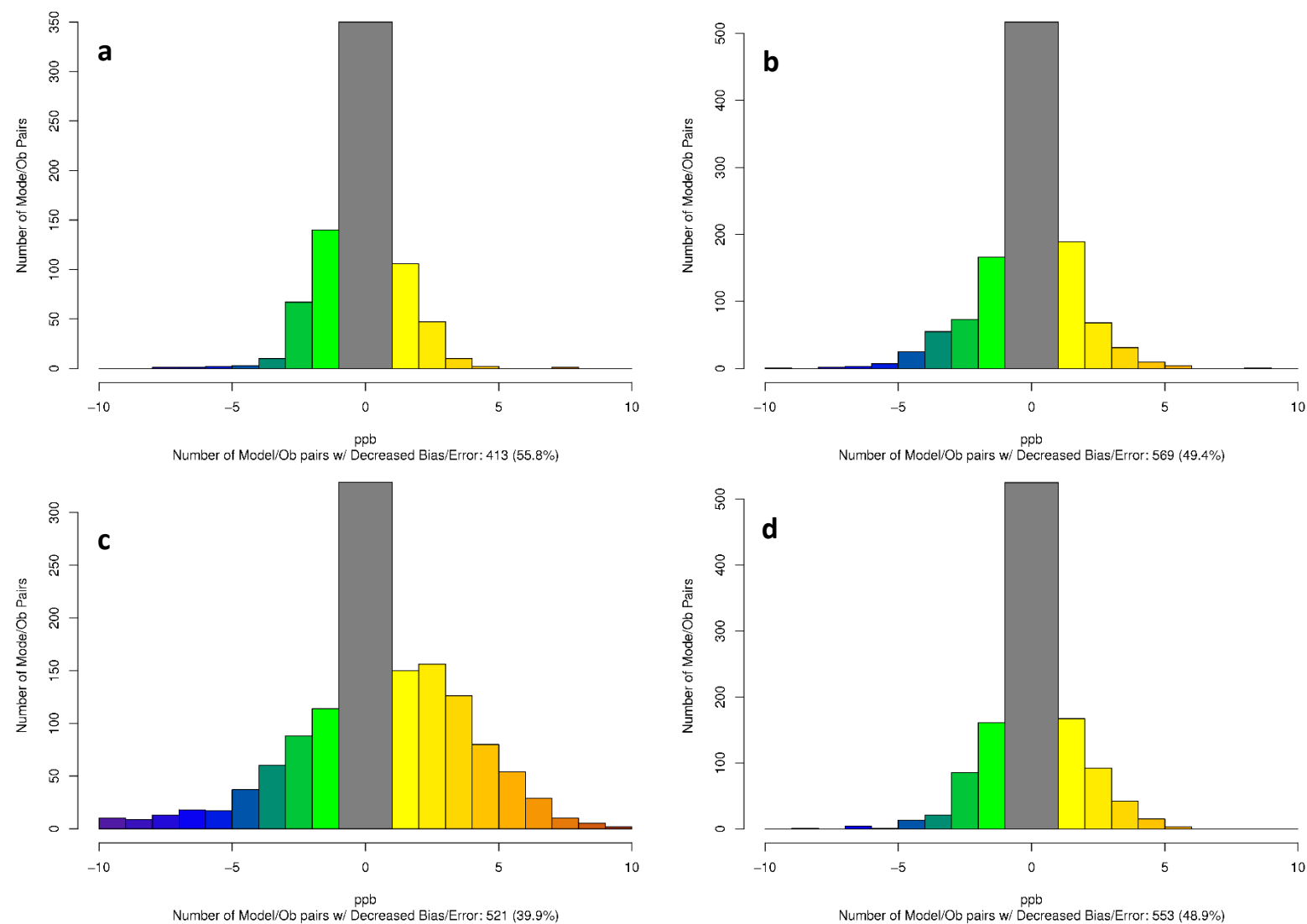


Figure S9: Histograms of the difference in the absolute value of monthly average O₃ mean bias for winter (DJF; top left), spring (MAM; top right), summer (JJA; bottom left) and fall (SON; bottom right) between CMAQ v5.0.2_Base and v5.1_Base (CMAQv5.1_Base – CMAQv5.0.2_Base). All plots are in units of ppbV. Cool colors indicate a reduction in O₃ mean bias in CMAQv5.1_Base while warm color indicate an increase in O₃ mean bias CMAQv5.1_Base.

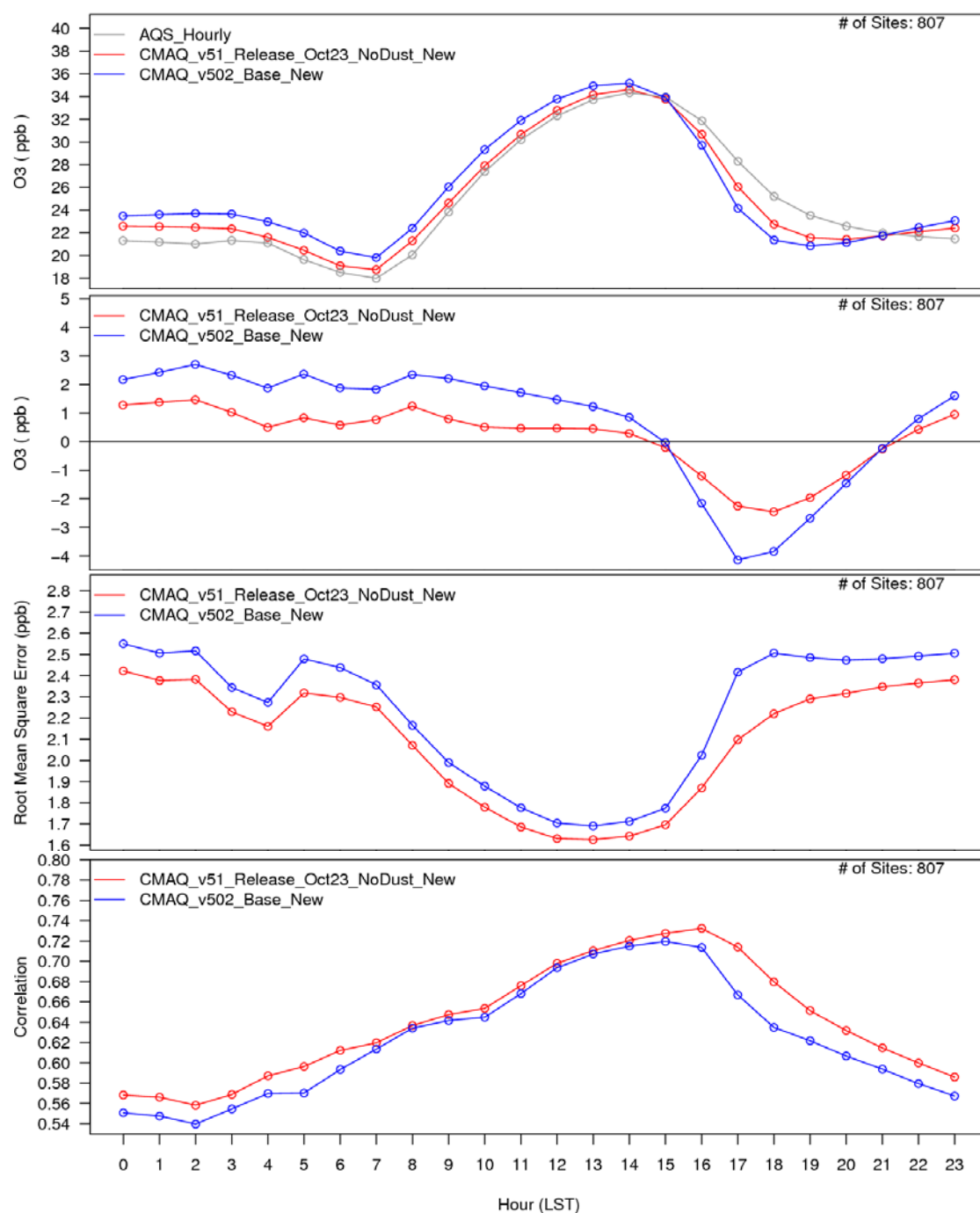


Figure S10: Diurnal time series of winter O₃ from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

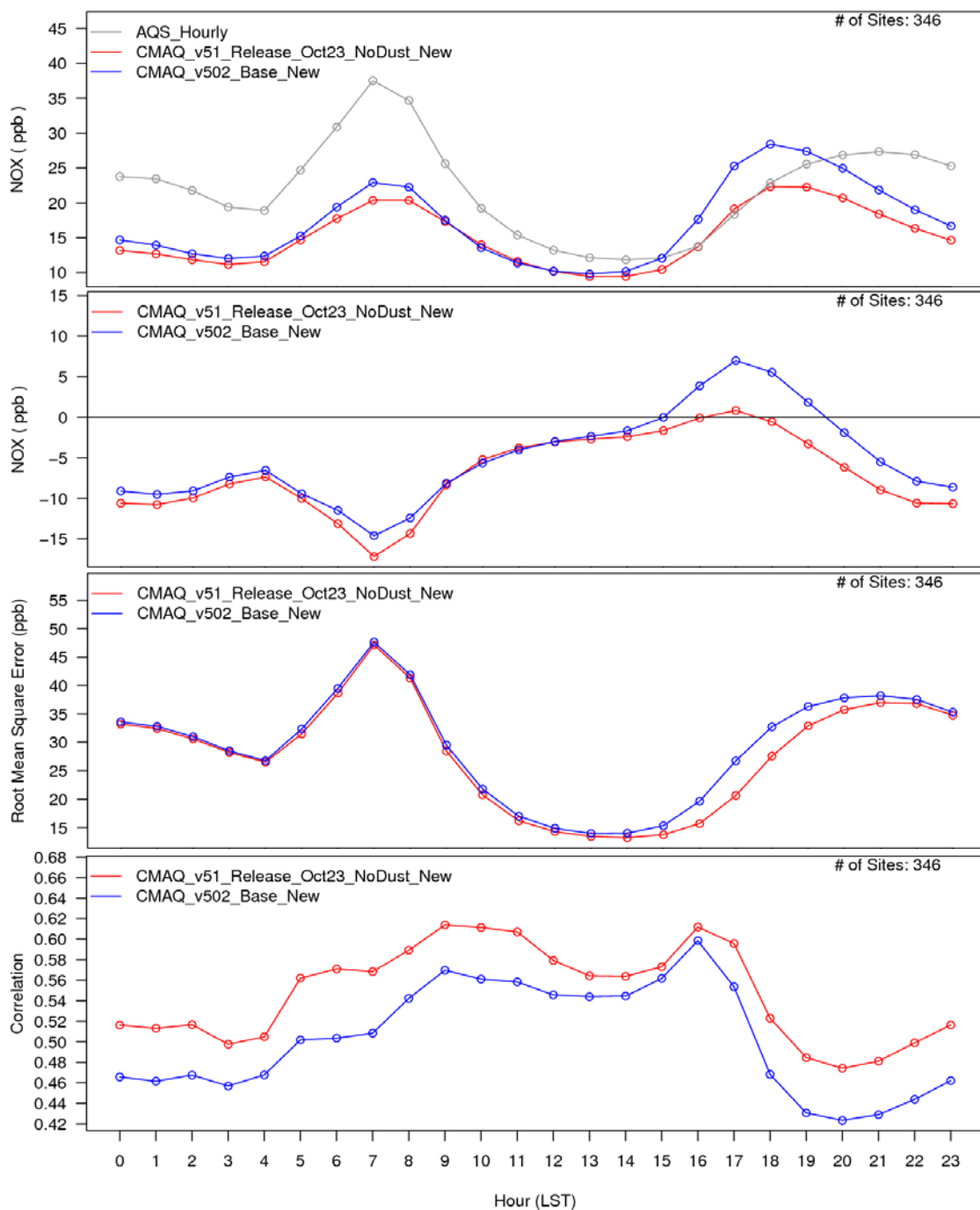


Figure S11: Diurnal time series of winter NO_x from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

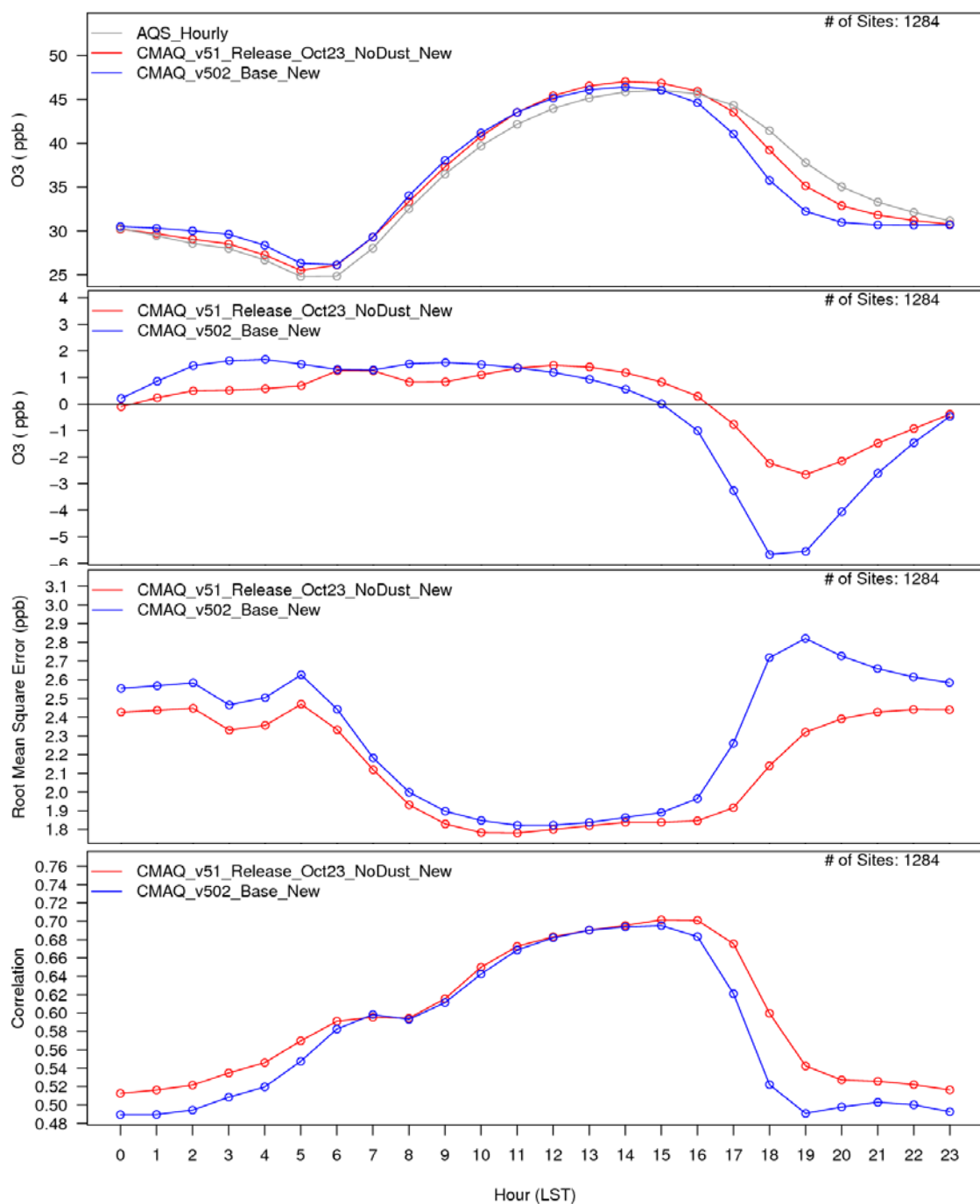


Figure S12: Diurnal time series of spring O₃ from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

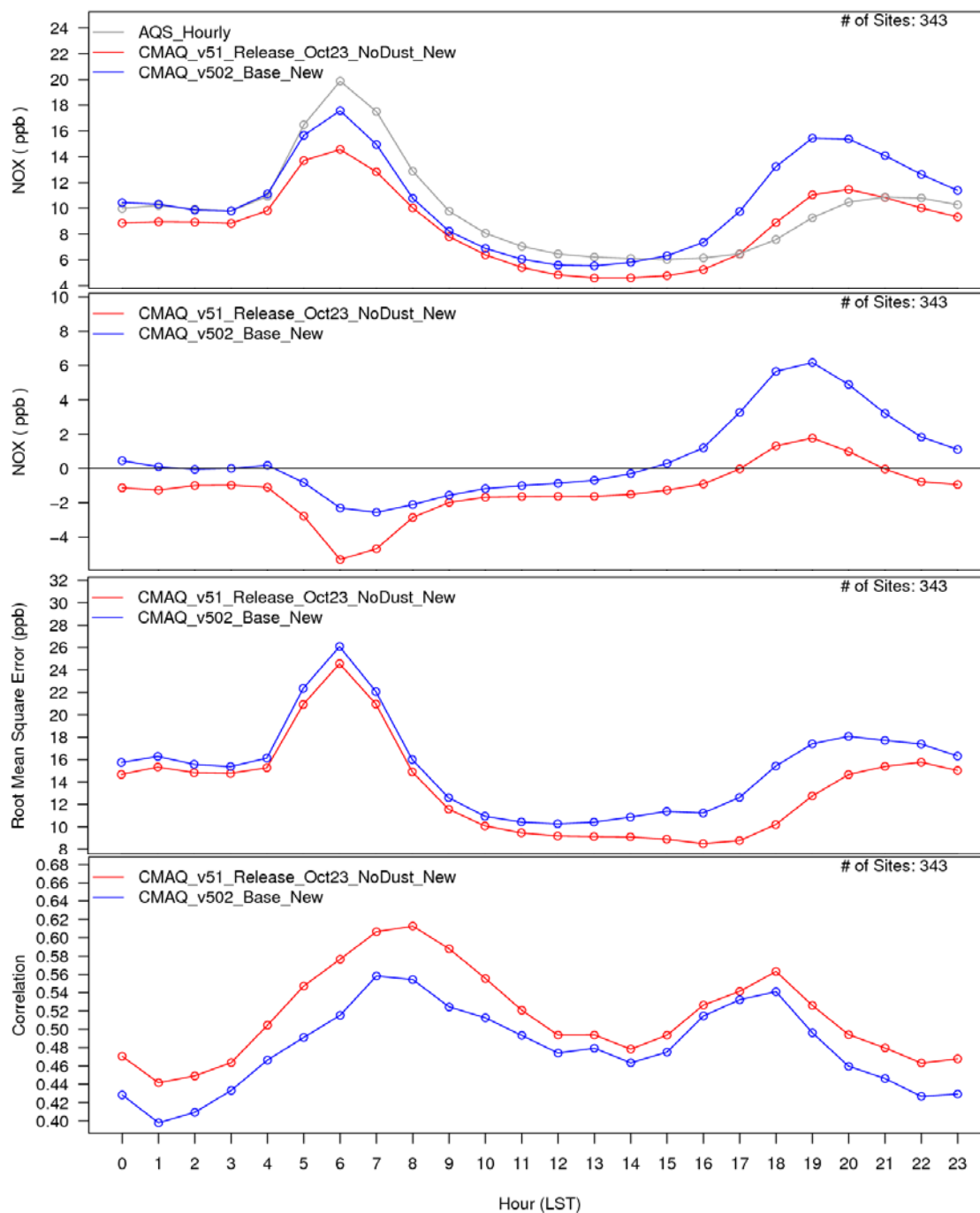


Figure S13. Diurnal time series of spring NO_x from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

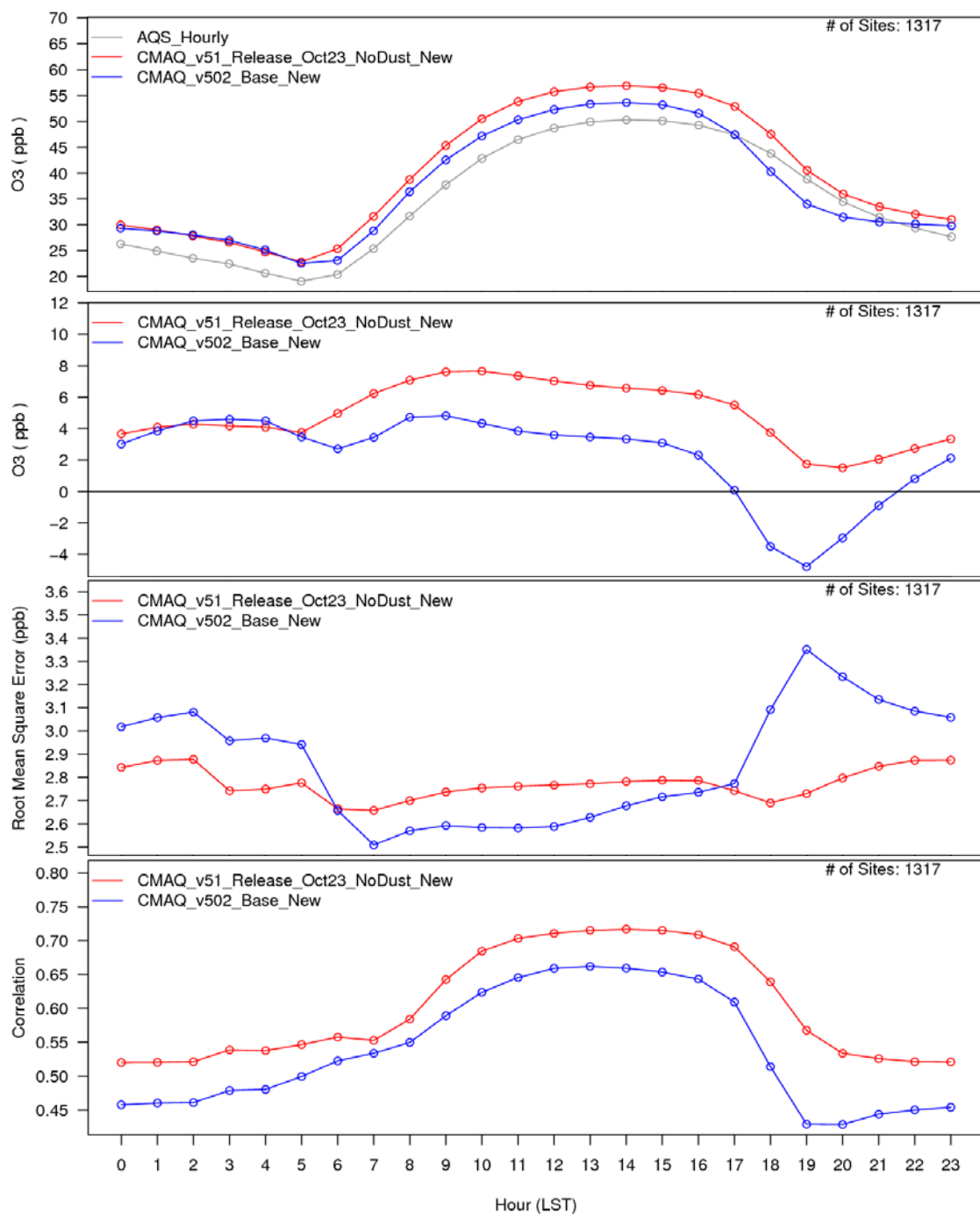


Figure S14: Diurnal time series of summer O₃ from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

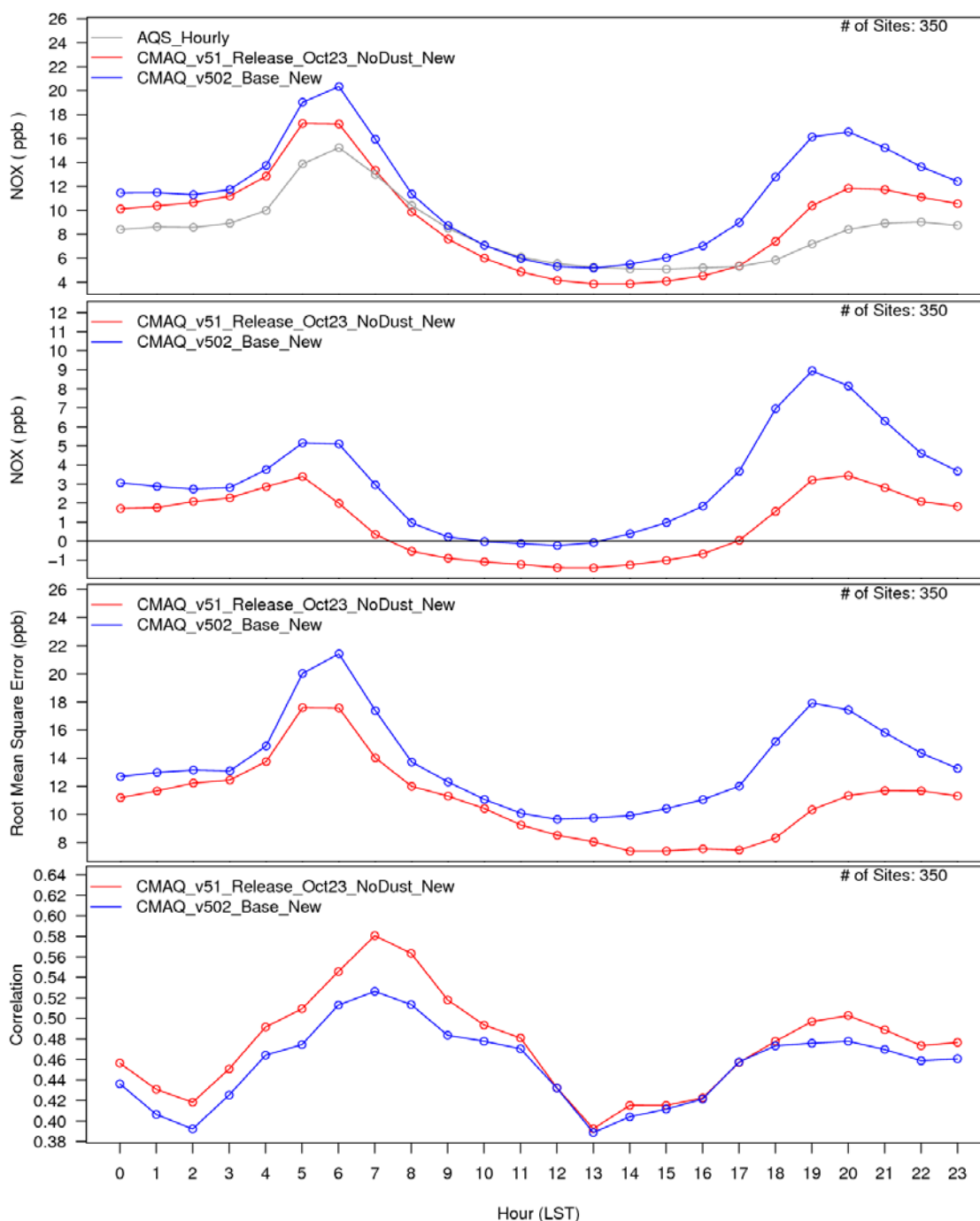


Figure S15: Diurnal time series of summer NO_x from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

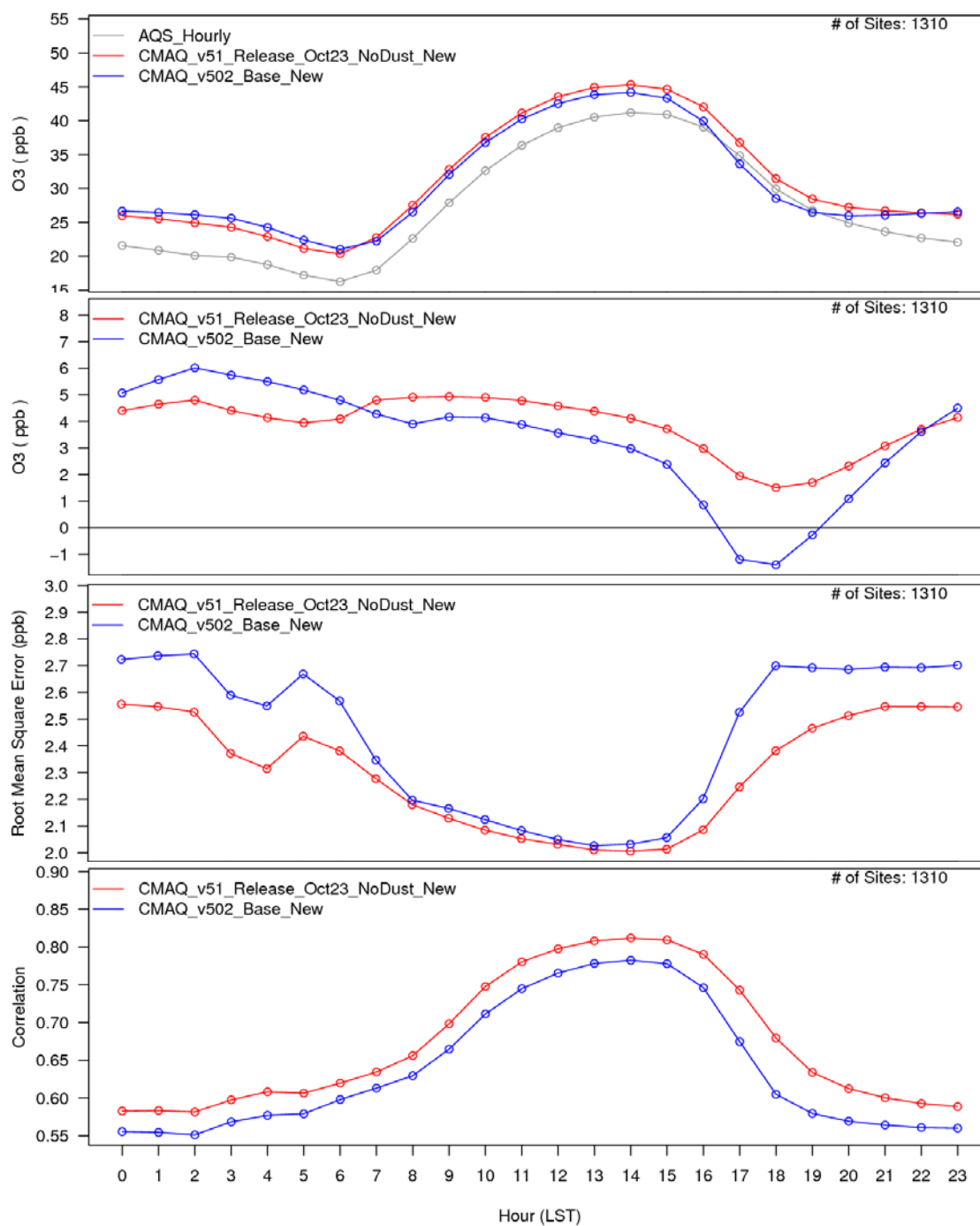


Figure S16: Diurnal time series of fall O₃ from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.

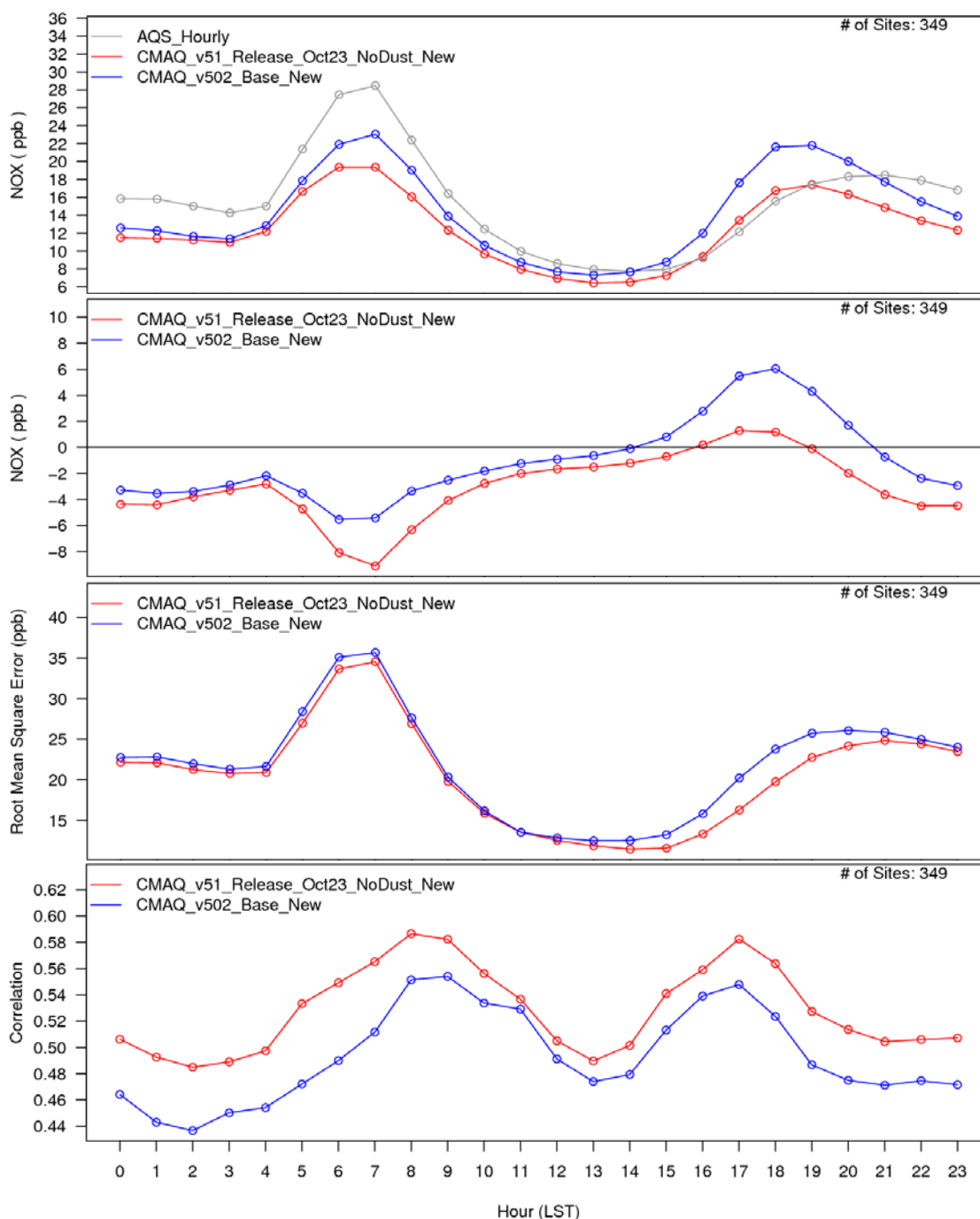


Figure S17: Diurnal time series of fall NO_x from AQS observations (grey), CMAQv5.0.2_Base (blue) and CMAQv5.1_Base (red) for concentration (top), mean bias (top middle), root mean square error (bottom middle) and correlation (bottom). All units are in ppbV except for correlation.