12 Feb. 2026

Dear Emmanouil Flaounas,

We provide below a complete copy of our responses to the two reviews of our Part 1 manuscript, including the review comments themselves (in black font), our responses (in blue font), and a description of the textual changes that we made to the manuscript as part of our response to the reviews (in blue font). New references have also been included (in red font).

In addition we made some minor editorial changes to the manuscript in the way we referred to specific and general versions of our forecast system. This was for consistency with our response to a comment by Reviewer 1 on our Part 2 manuscript.

We have also provided two separate PDF files, one containing our revised manuscript and one containing a marked-up version showing the changes that we have made. Please note that we did not make any changes to the Supplement to the manuscript that we submitted last September.

With best regards,

Michael Moran

Authors' Response to Reviewers of GMDD Manuscript "*Operational chemical weather forecasting with the ECCC online Regional Air Quality Deterministic Prediction System version 023 (RAQDPS023) – Part 1: System description*" by Moran et al. (2026)

Reviewer 1

General comments:

The manuscript provides a very comprehensive description of the RAQDPS023 modeling system used by Environment and Climate Change Canada to provide air quality forecasts to the public. As such, it is a rare example of a one-stop technical and scientific documentation that allows the reader to gain an understanding of the many different aspects that go into building such a system. It is also extremely well written and structured so that despite its length it is easy to follow. I commend the authors for the care they took in compiling the references that underly the scientific formulations of RAQDPS023. This extensive list of references in conjunction with the detailed descriptions of all RAQDPS023 science processes creates a rare repository of knowledge not only about RAQDPS023 but also about the tremendous amount of effort it takes to design, implement, and operationalize air quality modeling systems more generally. My specific comments listed below are minor and/or editorial in nature.

Response: We thank the reviewer for their generous assessment of this manuscript and for their constructive comments. We respond below (in blue font) to their specific comments.

Specific comments:

Page 3: "Schiermeir, 1978" should be "Schiermeier, 1978". While the doi of the scanned copy available through the ACS legacy archives (https://pubs.acs.org/doi/abs/10.1021/es60142a608) indeed mis-spelled the author's last name, the actual scanned copy available under that doi correctly shows the last name as "Schiermeier"

Response: Corrected in text and in References section.

Page 7, line 218 and Table 1: Consider using "horizontal domain size" instead of "horizontal grid size" since the latter could potentially be misinterpreted to refer to the size (spacing) of individual grid cells which is identical between RAQDPS023 and RDPS 8.0.0.

Response: Made several changes as suggested.

Page 8, line 255: Maybe change "included a small number of meteorological tracers" to "included two meteorological tracers (water vapour and cloud water)"

Response: Made this change as suggested.

Page 8, line 266: Insert "and" between "meteorological" and "chemical"

Response: Corrected.

Page 14, lines 454 - 456: Please clarify relative to which starting point (e.g., 10 bin GEM-MACH configuration, RAQDPS022) this modification of the numerical solution was implemented.

Response: This treatment was implemented early in the development of the RAQDPS. The second half of this sentence has been revised as follows:
"... the numerical solution of these processes was modified for the simplified two-bin description of the PM size distribution **early in the development of the RAQDPS** to reduce numerical errors"

Page 15, line 476: Consider changing "where" to "whereas"

Response: Corrected.

Page 15, lines 495 – 496: This question reveals my lack of understanding of aerosol schemes, but I was still curious what "partially activated" refers to. If there is a critical particle radius above which all aerosol particles are activated as stated on line 489, and if the two bins each represent aerosols of a discrete size, how does partial activation occur? Does each discrete size bin assume an internal distribution of particle radii, allowing a determination of the fraction of particles in a bin that exceed the critical radius above which aerosols are activated?

Response: "Partially activated" refers to a portion (a fraction) of the particles in a size bin being activated. The possibility of partial activation in this aerosol activation scheme is a consequence of

the critical particle radius for activation falling between the lower and upper boundaries of a size bin. We have revised the text in two places to try to provide a clearer explanation.

In Sect. 3.3.2 we have added some detail about $N_{p,i}$, the particle number concentration in size bin i:

"The overall condensation rate to a given particle size bin also depends on the particle number concentration for that bin **($N_{p,i}$). This quantity is calculated as the total dry volume of the eight PM chemical components in that bin (i.e., excluding aerosol water) divided by the volume of a spherical particle whose diameter is the arithmetic mean of the size boundaries of that bin.** Note that condensation to multiple size bins will occur simultaneously."

And in Sect. 3.3.5 we have revised the last sentence as follows:

"Note that **one consequence of this scheme is that only some of the particles in a size bin may be activated when the critical particle radius for activation falls between the lower and upper boundaries of a size bin**. For the RAQDPS023 with only two size bins, **all of the particles in** the PM_{cf} size bin **(i.e., $N_{p,2}$)** must be activated **before any particles in** the fine size bin can be activated."

Page 16, line 521: Would H2O (water vapour) profiles not be available from GEM?

Response: H2O (water vapour) profiles are available at every time step from GEM as suggested by the reviewer, and they are used by the gas-phase chemistry mechanism. The intent of this sentence was to state that five gas-phase species are needed by the ADOM-2 mechanism as reactants but are not treated as products. This means that they are not changed by gas-phase chemistry, thus reducing the computational burden required to apply the gas-phase chemistry operator. To make this clearer the sentence has been modified as follows:

"Note from Table 3 that the abundances of **42** of these **model** gas-phase species were forecast and advected while the abundances of **four model** species (CH4, C2H6, **O2**, M) had specified, time-invariant vertical profiles and were not advected **or modified by gas-phase chemistry but were considered as reactants in some of the ADOM-2 reactions (Table 4), as was H2O, the water vapour field from GEM (huplus; see Table 2).** "

Page 16, lines 537 – 539: Maybe comment on the implications for SOA formation when omitting emissions of organic acids and approaching the decision of which emissions to retain solely from a reactivity point of view.

Response: This is a good point. The following sentence has been added after line 539 to address this point:

"While the omission of these categories is defensible from a reactivity perspective, the neglect of organic acids in particular may remove a potential source of secondary organic aerosol (e.g., Makar et al., 2003b; Fisseha et al., 2004)."

As well one sentence was modified in Sect. 6.2 to expand on the issue of potential SOA sources that are not yet considered:

"Some of these schemes also include oxidation pathways for SOA formation in cloud droplets, **such as organic acids, glyoxal, and isoprene epoxydiols** (e.g., Gong et al., 2011; **Marais et al., 2016; Fahey et al., 2017; Lamkaddam et al., 2021; Luu et al., 2025**)."

Fahey, K. M., Carlton, A. G., Pye, H. O. T., Baek, J., Hutzell, W. T., Stanier, C. O., Baker, K. R., Appel, K. W., Jaoui, M., and Offenberg, J. H.: A framework for expanding aqueous chemistry in the Community Multiscale Air Quality (CMAQ) model version 5.1, Geosci. Model Dev., 10, 1587–1605, https://doi.org/10.5194/gmd-10-1587-2017, 2017.

Fisseha, R., Dommen, J., Sax, M., Paulsen, D., Kalberer, M., Maurer, R., Höfler, F., Weingartner, E., and Baltensperger, U.: Identification of organic acids in secondary organic aerosol and the corresponding gas phase from chamber experiments, Anal. Chem., 76, 6535–6540, https://doi.org/10.1021/ac048975f, 2004.

Lamkaddam, H., Dommen, J., Ranjithkumar, A., Gordon, H., Wehrle, G., Krechmer, J., Majluf, F., Salionov, D., Schmale, J., Bjelić, S., Carslaw, K. S., El Haddad, I., and Baltensperger, U.: Large contribution to secondary organic aerosol from isoprene cloud chemistry, Sci. Adv., 7, eabe2952, https://doi.org/10.1126/sciadv.abe2952, 2021.

Luu, R., Schervish, M., June, N. A., O'Donnell, S. E., Jathar, S. H., Pierce, J. R., and Shiraiwa, M.: Global simulations of phase state and equilibration time scales of secondary organic aerosols with GEOS-Chem, ACS Earth Space Chem., 9, 288–302, https://doi.org/10.1021/acsearthspacechem.4c00281, 2025.

Marais, E. A., Jacob, D. J., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Krechmer, J., Zhu, L., Kim, P. S., Miller, C. C., Fisher, J. A., Travis, K., Yu, K., Hanisco, T. F., Wolfe, G. M., Arkinson, H. L., Pye, H. O. T., Froyd, K. D., Liao, J., and McNeill, V. F.: Aqueous-phase mechanism for secondary organic aerosol formation from isoprene: application to the southeast United States and co-benefit of SO_2 emission controls, Atmospheric Chem. Phys., 16, 1603–1618, https://doi.org/10.5194/acp-16-1603-2016, 2016.

Page 26, line 877: typo, change "haf already" to "had already"

Response: Corrected.

Page 27, line 898: Could you elaborate on this additional information and how it was used to adjust the inventories?

Response: A description of this additional information and how it was used to adjust the inventories is provided in the first part of Sect. S2. Lines 897-898 have been modified slightly to point to this section:

"**As described in Sect. S2 the** first step was to make some adjustments to the Canadian and U.S. inventories to account for additional **and missing** information."

Page 28, line 942: Should this be "the usual hourly anthropogenic and biogenic emissions" instead of just "the usual hourly anthropogenic emissions"?

Response: Yes, other natural emissions should be mentioned here. The latter part of the sentence has been modified as follows:

"… except for the addition of hourly BB emissions to the usual hourly anthropogenic emissions **and the hourly biogenic and sea-salt emissions described in the next section** (Pavlovic et al., 2016)."

Page 30, lines 995 – 1001: Was there any dependency of soil NO emissions on precipitation to represent the pulsing effect described in the Yienger-Levy framework?

Response: No, there was not. This was another limitation of BEIS 3.09, and BEIS was modified to account for the Yienger-Levy framework in BEIS 3.10, 3.11, and 3.12 (see https://www.epa.gov/air-emissions-modeling/biogenic-emission-inventory-system-beis). The following sentence has been appended to line 1008 to note this limitation:

"One remaining limitation, however, that was addressed in later versions of BEIS was that soil NO emissions did not depend on soil moisture or precipitation (cf. Yienger and Levy II, 1995)."

Page 33, line 1111: Please check if "for 900 s" is needed here. With the way the end of the sentence is written ("for 900 s for three shorter 300 s time steps"), I am not sure if the only point of the sentence is to say that a single MACH chemistry execution every 900 s still took four times longer than the combined time it took to execute three GEM 300 s physics time steps, or if there is some additional information being conveyed here.

Response: No additional information is being conveyed. Rather, the intent was to ensure clarity, but as suggested by the reviewer the mention of "for 900 s" here is likely unnecessary and has been removed.

Page 36, lines 1200 – 1211: It might be good to provide a brief summary of which emission inputs (anthropogenic and natural) were used in these MOZART-4 simulations. Did the anthropogenic emissions represent 2009 conditions? Were aircraft, lightning NO, and soil NO emissions considered? Adding such information could set the stage for the updates to LBC in RAQDPS025 and future versions discussed in later sections of the manuscript.

Response: This is a good suggestion. The following text has been inserted at line 1204:

"Global anthropogenic emissions of 20 model species were obtained from the ARCTAS inventory (Streets et al., 2006; D'Allura et al., 2011; http://bio.cgrer.uiowa.edu/arctas/emission.html; https://www.acom.ucar.edu/gctm/mozart/subset) and biogenic, biomass burning, marine, lightning, and volcanic emissions were also considered (Emmons et al., 2010)."

Streets, D. G., Zhang, Q., Wang, L., He, K., Hao, J., Wu, Y., Tang, Y., and Carmichael, G. R.: Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations, J. Geophys. Res. Atmospheres, 111, 2006JD007118, https://doi.org/10.1029/2006JD007118, 2006.

D'Allura, A., Kulkarni, S., Carmichael, G. R., Finardi, S., Adhikary, B., Wei, C., Streets, D., Zhang, Q., Pierce, R. B., Al-Saadi, J. A., Diskin, G., and Wennberg, P.: Meteorological and air quality forecasting using the WRF–STEM model during the 2008 ARCTAS field campaign, Atmos. Environ., 45, 6901–6910, https://doi.org/10.1016/j.atmosenv.2011.02.073, 2011.

Pages 45, lines 1493 – 1503: Given the episodic nature of processes affecting large-scale distributions of O3, CO, and PM2.5, why did the LBC updates in RAQDPS025 still adopt an approach based on climatology? Were there any differences in the types of anthropogenic and natural emissions considered in the MOZART-4 vs. CAM-chem simulations?

Response: The reviewer raises a very important issue here. They are correct that time- and space-dependent chemical LBCs are preferable to climatological CLBCs in principle. However, the

challenge for an operational forecasting application is that such time- and space-varying chemical LBCs depend on the availability of global chemical weather forecasts for multiple days in real time, which either requires an in-house operational global chemical weather model or else close cooperation and dependence on an external agency with an operational global chemical weather model. Even if such an external partner could be found, dependence on an external agency introduces high risk for an operational system. An in-house global version of GEM-MACH has in fact been under development at ECCC for some time but is not yet ready for deployment, which leaves climatological CLBCs as the best available solution in the meantime.

The new climatological CLBCs based on eight years of CAM-chem simulations that are briefly described in Sect. 6.1.5 are improvements over the MOZART-4-based CLBCs in at least four ways. First, they are more truly "climatological" in that they were calculated using eight years of simulations rather than a single year, where that single year might be an outlier. Second, they have higher temporal resolution: monthly vs. seasonal. Third, they are based on results from a newer CTM that employs an updated and more complete representation of atmospheric chemistry. And fourth, they are based on a newer multi-year anthropogenic global emissions inventory and consider aeolian dust emissions.

The following text has been appended to Sect. 6.1.5 to describe the CAM-chem emissions and to address the issue of time- and space-varying CLBCs:

"Year-specific anthropogenic emissions for these annual simulations were obtained from CAMS-GLOB-ANT v5.1, a recent multi-year global emissions inventory (Soulie et al., 2023, 2024) and natural emissions from biogenic, biomass burning, marine, lightning, dust, and volcanic sources were also considered (Emmons et al., 2020).

While such climatological CLBCs do not address the episodic nature of the large-scale distributions of longer-lived pollutants such as O_3, CO, and $PM_{2.5}$, they can still be broadly representative of typical chemical inflows. The alternative is to supply time- and space-varying CLBCs based on forecasts from a compatible global chemical weather model. Such a system is currently under development at ECCC, but it is not yet ready for operational deployment."

Soulie, A., Granier, C., Darras, S., Zilbermann, N., Doumbia, T., Guevara, M., Jalkanen, J.-P., Keita, S., Liousse, C., Crippa, M., Guizzardi, D., Hoesly, R., and Smith, S. J.: Global anthropogenic emissions (CAMS-GLOB-ANT) for the Copernicus Atmosphere Monitoring Service simulations of air quality forecasts and reanalyses, ECCAD-AERIS, 2023.

Soulie, A., Granier, C., Darras, S., Zilbermann, N., Doumbia, T., Guevara, M., Jalkanen, J.-P., Keita, S., Liousse, C., Crippa, M., Guizzardi, D., Hoesly, R., and Smith, S. J.: Global anthropogenic emissions (CAMS-GLOB-ANT) for the Copernicus Atmosphere Monitoring Service simulations of air quality forecasts and reanalyses, Earth Syst. Sci. Data, 16, 2261–2279, https://doi.org/10.5194/essd-16-2261-2024, 2024.

Page 46, lines 1520 – 1522: Does this update eliminate any dependence on the five broad phenological seasons described in the last paragraph of Section 3.9?

Response: This update reduces that dependence, and it also introduces longitudinal and altitudinal variations for LAI. However, other important parameters in the dry deposition parameterization (e.g., z_0, R_{cut}, R_{gd}, R_{can}, R_{exp}, R_{st_min}) are still based on lookup tables and hence are dependent on those phenological seasons (Makar et al., 2018b). A similar limitation was noted in

lines 1561-1562 for the updated calculation of biogenic emissions. In principle it might be possible to diagnose location-dependent phenology from the monthly LAI climatology to reduce the current strong latitude dependence. This possibility is noted in Sect. 6.2.

However, as this is an important limitation of the current gas-phase dry deposition scheme, the following text has been inserted in line 767 in Sect. 3.9 to explain the need for the simplified phenological seasons:

"These seasons are used in lookup tables to specify seasonal values of a number of required parameters, including soil, cuticle, canopy, and minimum stomatal resistances, for 15 land-use types (Makar et al., 2018b)."

Another sentence has been added after line 1522 that should raise and answer the reviewer's question for other readers:

"The use of monthly LAI fields reduces but does not remove the dependence on five phenological seasons that was described in Sect. 3.9."

Reviewer 2

General comments:

This manuscript presents the operational chemical weather forecasting system of ECCC. I would like to express my applause for the authors' many works to summarize this. This will be beneficial to trace the entire history of modeling development in ECCC, and we can learn from this broad perspective in this manuscript. Therefore, it's unavoidable that it ends up being long. I do not have any critical concerns about its current presentation quality, but I have minor requests for improving the readability.

Response: We thank the reviewer for their generous assessment of this manuscript and for their helpful comments. We respond below (in blue font) to their specific comments.

Specific comments:

Abstract (after line 30): Because the Part 2 manuscript presents the modeling performances not only for PM2.5 but also for gases and depositions, it would be better to provide this relevant information, such as ADOM-2 gas-phase chemistry. The current description just relies on the aerosol.

Response: Thank you for this comment. We have modified lines 34–37 of the Abstract to include information about the gas- and aqueous-phase chemical schemes.

"Details covered in this paper include a summary of the dynamical representations and physical parameterizations used in the three GEM-based forecast systems, which are highly harmonized, the chemical **species and** parameterizations used in the MACH chemistry module, **including gas-phase, aqueous-phase, and inorganic heterogeneous schemes and associated** numerical solvers, system inputs, including both anthropogenic and natural emissions of chemical species,

system outputs, and run configuration, strategies, and timings. One simplification **in addition to the use of the condensed ADOM-2 gas-phase chemistry scheme that was made** to reduce RAQDPS023 execution time for operational deployment was to …"

Lines 967-968: I understand that these emissions were not considered in RAQDPS023, but is there a rationale for excluding them from the forecasting system in Canada? Regarding the aerolian dust, I could see the discussion in the final paragraph of Section 3.12; what about other sources?

Response: We agree with the reviewer that a one-sentence mention of model simplifications with respect to natural emissions is insufficient. We have moved this sentence to the end of Section 3.11.3 and expanded it to give a rationale as follows:

"Note, however, that some other sources of natural emissions, including lightning emissions, volcanic emissions, pollen and other biological emissions, **oceanic gas-phase emissions**, and aeolian dust emissions, were not considered by the RAQDPS023. While all of these emissions affect atmospheric chemistry, their impacts on near-surface gas-phase and $PM_{2.5}$ concentrations over Canada are assumed to be small, especially when compared to biogenic, biomass burning, and sea-salt emissions, the three types of natural emissions that are considered. Lightning NO emissions occur intermittently above the Earth's surface. While they have little influence on surface concentrations locally, they can influence NO_3^- wet deposition (e.g., Appel et al., 2011; Zhang et al., 2018b). Volcanic SO_2 emissions can affect atmospheric concentrations and deposition, but active volcanoes are not a significant source in populated areas of Canada or the U.S. although this is not true in Mexico (e.g., Fioletov et al., 2016). Pollen and other biological PM emissions are an important source of atmospheric PM and aeroallergens, but they occur mainly as particles larger than 2.5 μm in diameter, including pollen grains generally larger than 20 μm but fungal spores in the 1–10 μm range (e.g., Efstathiou et al., 2011; Sierra-Heredia et al., 2018; Subba et al., 2021). Oceanic emissions of gas-phase species such as DMS and isoprene contribute to background atmospheric chemistry, although DMS emissions are small at higher latitudes and marine isoprene emissions are small compared to terrestrial emissions (e.g., Bates et al., 1992; Khan et al., 2025). However, as discussed in Sect. 6.2 emissions of halogen species such as iodine can affect surface ozone levels. Aeolian dust emissions are discussed further in Sect. 3.12, but such emissions are very sporadic and are much less important in Canada than the U.S."

Appel, K. W., Foley, K. M., Bash, J. O., Pinder, R. W., Dennis, R. L., Allen, D. J., and Pickering, K.: A multi-resolution assessment of the Community Multiscale Air Quality (CMAQ) model v4.7 wet deposition estimates for 2002–2006, Geosci. Model Dev., 4, 357–371, https://doi.org/10.5194/gmd-4-357-2011, 2011.

Bates, T. S., Lamb, B. K., Guenther, A., Dignon, J., and Stoiber, R. E.: Sulfur emissions to the atmosphere from natural sources, J. Atmospheric Chem., 14, 315–337, https://doi.org/10.1007/BF00115242, 1992.

Efstathiou, C., Isukapalli, S., and Georgopoulos, P.: A mechanistic modeling system for estimating large-scale emissions and transport of pollen and co-allergens, Atmos. Environ., 45, 2260–2276, https://doi.org/10.1016/j.atmosenv.2010.12.008, 2011.

Fioletov, V. E., McLinden, C. A., Krotkov, N., Li, C., Joiner, J., Theys, N., Carn, S., and Moran, M. D.: A global catalogue of large SO2 sources and emissions derived from the Ozone Monitoring Instrument, Atmospheric Chem. Phys., 16, 11497–11519, https://doi.org/10.5194/acp-16-11497-2016, 2016.

Khan, M. A. H., Holland, R., Mould, C., Bacak, A., Percival, C. J., and Shallcross, D. E.: Isoprene emissions, oxidation chemistry and environmental impacts, Atmosphere, 16, 259, https://doi.org/10.3390/atmos16030259, 2025.

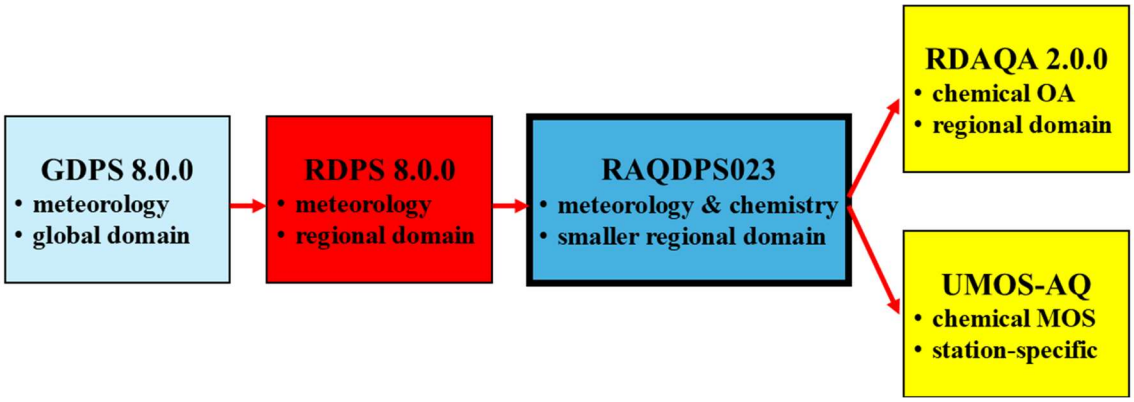
Sierra-Heredia, C., North, M., Brook, J., Daly, C., Ellis, A. K., Henderson, D., Henderson, S. B., Lavigne, É., and Takaro, T. K.: Aeroallergens in Canada: Distribution, public health impacts, and opportunities for prevention, Int. J. Environ. Res. Public. Health, 15, 1577, https://doi.org/10.3390/ijerph15081577, 2018.

Subba, T., Lawler, M. J., and Steiner, A. L.: Estimation of possible primary biological particle emissions and rupture events at the southern Great Plains ARM site, J. Geophys. Res. Atmospheres, 126, e2021JD034679, https://doi.org/10.1029/2021JD034679, 2021.

Zhang, Y., Mathur, R., Bash, J. O., Hogrefe, C., Xing, J., and Roselle, S. J.: Long-term trends in total inorganic nitrogen and sulfur deposition in the US from 1990 to 2010, Atmospheric Chem. Phys., 18, 9091–9106, https://doi.org/10.5194/acp-18-9091-2018, 2018b.

Line 3290 (Figure 1): I would like to request an improvement in this figure. For example, RAQDPS includes various sub-components, and it could be included within this figure. As the final application (post-processing), RDAQA and UMOS-AQ are described, but the purposes for each part could be briefly described in the arrow. Moreover, the colors for RDPS and RAQDPS could be unified in Figure 2 for better presentation.

Response: As suggested by the reviewer we have revised this figure to add more information about key characteristics of each system. We have also made the colours for the RDPS and RAQDPS boxes consistent with the colours used in Fig. 2 for the corresponding system grids.



Technical points:

Lines 321, 335, 910, 916, 1150, 1380, 1528, 1531, 1604: For Moran et al. (2025), it will be better to represent this as “a companion paper by Moran et al. (2025)” (Line 52).

Response: This is a good suggestion, and we have made changes in a number of places in the paper to adopt it. We have also made similar changes in the Part 2 paper to refer to this paper.

Line 520: Use a subscript for the chemical species shown in this line.

Response: We have clarified the meaning of “species” in this sentence, which refers to the model gas-phase species listed in Table 3, by making the following modifications (in bold):

“… abundances of 42 of these **model** gas-phase species were forecast and advected while the abundances of four **model** species (CH4, C2H6, **O2**, M) had …”.

Line 577: Use subscript “NH3”.

Response: Made change as suggested.

Line 3263: Please define “LRT”.

Response: “LRT” is defined in the footnotes to Table 7. However, we have reordered these footnotes for clarity so that we first define L, R, and T, and we have also modified the nomenclature slightly by changing “LRT” to “$\mathbf{L_w}RT$”, in order to avoid confusion with the use of the symbol L (for liters) in the units of the universal gas constant R.