We thank Reviewer 1 and Reviewer 2 for their comments and suggestions. These have resulted in a considerable reworking of the original material, which, we hope, addresses their concerns. The updates have resulted in a much more "streamlined" version of the original manuscript, more directly focused on the final configuration of our results, and more clearly states their significance, in many cases through revised Figures and additional discussion.

In the following responses, the Reviewer comments have been repeated for reference in *italics*, and our point-by-point responses follow in regular font.

Reviewer 1:

Summary

This manuscript describes work to improve process representation of SO2 dry deposition in the GEM-MACH regional air quality model. Dry deposition is an important process to capture in chemistry transport models, but process realism is difficult to achieve due the sub-grid scale nature of the contributing processes. The authors address this problem by targeting dry deposition on wet vegetation surfaces. They expand GEM-MACH's dry deposition mechanism to include a scheme that calculates pH on the thin films of water that accumulate on foliage. They hypothesize that this will improve SO2 dry deposition in regions like the Athabasca Oil Sands, where high atmospheric concentrations of acidic and alkaline gases and aerosols can influence foliage pH, and therefore deposition of acidic and basic gases. The authors find that GEM-MACH can better predict SO2 dry deposition velocities in the Athabasca Oil Sands Region when this scheme is included, although the impacts on atmospheric SO2 concentrations is small.

We note that the impact on atmospheric SO2 concentrations can sometimes be large, depending on the amount of base cations on the foliage: for example, at HRZN station in June, the previous positive bias of 57.6% became 5.7% (a factor of 10 reduction in NMB), see revised Figure One of the Reviewer's other suggestions, regarding amalgamating some of the submitted manuscript Figures by month, also makes it clear that the pH dependence results in changes in average SO2 concentration and SO2 fluxes of up to a factor of two across our model domain (see responses below) – this is a significant perturbation. The impacts on atmospheric SO2 concentrations can sometimes be large, based on the Taylor plots; as we have mentioned in the revised text (see detailed comment responses).

Overall, the manuscript is well written in clear language. However, I think that the manuscript is over-long relative to the limited scope of the results. It is unclear what the wider impacts, if any, the new foliage water pH scheme would have on e.g. deposition

velocities of other acidic and basic gases like NH3 or HNO3 and what impact it might have a dusty or wildfire affected region.

While expanding the analysis to other species is a topic for future work, note that in the revised manuscript we have emphasized the role of wildfires as a source of base cation deposition in several places (revised manuscript Abstract, Introduction lines 84, Section 2 line 231, Section 5.2 discussion on Figures 6-9, Conclusions; search for "fire" in the revised manuscript). As mentioned in the revised Section 5.2, much of the increased deposition velocity going from May (Figure 6) to June and July (Figures 7,8) in the portion of the model domain associated with the boreal forest (upper right corner half of these Figures) is due to forest fire emissions and subsequent deposition of base cations.

While I recommend the manuscript for publication, I have some General and Technical comments below that should be addressed first.

General Comments

1. *Overall the manuscript is long relative to the scope of the results presented in Section5.2-5.4. In particular, I think that the length of Sections 3 and 4 detracts from the study's results. I suggest that the technical details of the GEM-MACH dry deposition scheme presented in Sections 3 and 4 are moved to the Supplementary Material (or possibly as an Appendix)*.

   We agree that the submitted sections 3 and 4 were unnecessarily lengthy. However, much of our work was in determining the most realistic approach for simulating the effects of pH on deposition, which is described in those sections, and we feel is therefore important for the readers of the article. Therefore, instead of moving the sections entirely, we put considerable effort into "streamlining" these sections, following the Reviewer's suggestions. Specifically:

   - Much of the background material of section 3 was moved to the Supplement material as suggested by the Reviewer (this material appears in the revised manuscript's Supplement, Appendix A).
   - Table 1 was moved to the revised Supplement, Table S1
   - The material describing mathematical details of the means in which the cuticle portion of the deposition flux was tracked and accumulated were moved to the Supplement, where they now appear as Appendix B and Appendix C. The main text now includes only the final two formulae resulting from that analysis and associated text, in Sections 4.3.1 and 4.3.2.
   - The text associated with the tests of both the simple charge balance adjustment (charge_adj) and the lower water layer threshold (Lw_low) were removed, in

favour of brief statements on the results of those tests, in the main document, with the resulting focus in Sect. 5 being on the results from the final configuration CALCCO3_Lw_high.

- As requested by the Reviewer in the detailed comments, we added an overview paragraph to Section 4. We also re-arranged the Section 4 presentation to better describe the portions of the algorithm required to simulate leaf cuticle pH in a more logical order, and harmonized symbols between these subsections, to make the Section have a more logical "flow" and be easier to follow. These subsection names follow from the introductory paragraph to Section 4 suggested by the Reviewer.

The resulting sections 3 and 4 have been reduced in size relative to those of the submitted manuscript by 26%. The numbered formulae presented in the main document has decreased from 41 to 17 equations. The revised sections are thus much more focused on the final version of the test system. We feel that the methodology has become much easier to follow as a result, and we thank the Reviewer for their suggestions and guidance.

The manuscript could then focus on relevant updates to GEM-MACH's dry deposition scheme since Makar et al., 2018 and the HETP scheme implemented to calculate foliage pH.

Agreed. This is what we have done in the revised manuscript.

2. *The manuscript could also be better focused by either summarizing Section 5.1 or moving it to the supplementary section. The author's find that: "This analysis underscores the role of $Lw\ Thres$ in determining the frequency and magnitude of high VdSO2 associated with elevated foliage pH is relatively minor." (L818-), which is useful, but including the comparison between the two Lw Thres schemes and the change_adj scheme detracts from the more interesting results from the more suitable CALCCO3_Lw_high scheme shown in Sections 5.2-5.4.*

We considerably reduced the size of section 5.1 by:

- Moving the submitted manuscript Figure 5 (comparing CALCCO3_Lw_high and CALCCO3_Lw_low) to the Supplement (now Figure S2). New Figure S2 is now mentioned in Section 4.4.
- Removing Table 2 and the associated discussion on the charge_adj_Lw_low/high and CALCCO3_Lw_Low test simulations, in favour of brief statements in Section 4 on the results of those simulations.

- Reducing Figure 6 from the submitted manuscript (Figure 5 in the revised manuscript) to show only the results from the CALCCO3_Lw_high simulations.
- Reducing the corresponding text significantly, to focus only on the bar chart comparison.

The resulting Section 5.1 is slightly over a page in length, compared to five pages in the submitted manuscript.

3. *I would also suggest that Section 5.4 follows Section 5.2 so that the impact of the interactive foliage pH on SO2 deposition velocity to deposition flux to atmospheric SO2 concentrations can be 'traced' along the dry deposition pathway.*

   The subsections of Section 5 were reorganized substantially in order to carry out the Reviewer's next comment below; the combination of the submitted manuscripts Figures 7, 8, 10 and 13 into the revised manuscript's Figures 6, 7, and 8. The text for the end of submitted manuscript section 5.1, as well as 5.2, 5.3, and 5.5 were therefore amalgamated into a new section 5.2 "Impact of pH on predicted SO2 deposition velocities, surface concentrations, and deposition fluxes", with the comparison to observations following in new section 5.3.

4. *It is difficult for the reader to interpret the spatial impact of the new foliage pH scheme on VdSO2, SO2 dry deposition flux and SO2 concentrations because the spatial plots for these quantities are shown in Figures 7, 8, 10 and 13. Would it be possible to combine some of these spatial plots to illustrate the cascading impact of the new foliage pH scheme. For example by plotting a single month (or multi monthly mean) for VdSO2, SO2 dry deposition flux and SO2 concentrations, possibly using multi-monthly means?*

   The submitted manuscript's Figures 7, 8, 10 and 13 were combined into the revised manuscript's Figures 6, 7, and 8, and Supplement Figures S3 -S6. The Reviewer had a good point here; the small size of the panels in the submitted manuscript figures was obscuring some of our results which were important to display clearly. The revised Figures 6, 7, and 8 show the May, June, July (respectively) values of surface pH in the AOSR region, and the corresponding ratios of the CALCCO3_Lw_high to Base Case values for the SO2 monthly average deposition velocity, surface concentration and deposition flux. With the larger panels it is much easier to see that the pH-dependence has resulted in changes in SO2 concentrations and SO2 deposition fluxes by more than a factor of two (and sometimes approaching a factor of 10), in each of these months. The foliage pH and the ratios of the model with the pH adjustment (CALCCO3_Lw_high) to the Base Case appear in the revised main

body of the text (for the monthly averages of the hourly SO2 deposition velocity, surface air concentration and deposition flux), and the Supplement contains the Base Case and CALCCO3_Lw_high maps used to generate the ratios. This makes the details in the ratio plots much easier to see, aiding in the description. The text for the end of submitted manuscript section 5.1, as well as its 5.2, 5.3, and 5.5 were therefore amalgamated into a new section 5.2, with the comparison to observations following in new section 5.3.

5. *There are instances where the text could be edited to eliminate repetition. For example L704-710 repetes details from Section 2. This would help to reduce the length of the manuscript.*

   The first few lines of section 5.1, which had previously read, *"As discussed in Sect. 2, the predicted foliage pH is a new driving parameter that modulates the dry deposition velocity of SO_2 in this work ($V_{d_{SO_2}}$), specifically by allowing a time-varying $H^*_{SO_2}$ in Eq. 5 and Eq. 7. As demonstrated in Fig. 2, a very small increase in the predicted foliage pH from the current GEM-MACH value of 6.68 can dramatically increase $V_{d_{SO_2}}$, but the pH impact on modulating $V_{d_{SO_2}}$ is most important only for a rather small pH range. For example, increasing the pH above 9.0 has little effect on further increasing $V_{d_{SO_2}}$ (i.e., as $r_{cut}(SO_2) \rightarrow 0$); likewise decreasing the pH below 5.0 has little effect further decreasing $V_{d_{SO_2}}$(i.e., as $r_{cut}(SO_2) \rightarrow \infty$). Furthermore, studies have indicated that the quantity of liquid water retained on the leaf, defined here in terms of the leaf water layer thickness, crucially affects the leaf water layer pH (Burkhardt and Eiden 1990; Chameides 1987; Klemm et al., 1987).",* have been shortened and revised to read, "In the above sections, we have shown that small changes in foliage pH are capable of modifying $\mathrm{V_{d_{SO_2}}}$ via time-varying $\mathrm{H^*_{SO_2}}$, with leaf water layer thickness as a key controlling parameter." The sentence "The quantity of liquid water retained on the leaf, defined here in terms of the leaf water layer thickness, crucially affects the leaf water layer pH (Burkhardt and Eiden 1990; Chameides 1987; Klemm et al., 1987)," has been moved to Section 4.3.

6. *In addition, the manuscript text repeats the figure captions in many cases, which is not necessary.*

   We see the Reviewer's point, having compared the Figure captions identified by the reviewer and the corresponding text. We've shortened the main text and expanded the Figure captions (where necessary; as the Reviewer pointed out, sometimes the information was being repeated).

Technical Comments

Abstract

1. *L37-38: Typo in "We examine here the potential the simultaneous…"*

Suggest "We examine here the potential for simultaneous…" Thanks, corrected.

Introduction

1. *Consider using sub sections (e.g., by key process - foliage surface pH, canopy particle deposition, leaf water dynamics, and dust-gas interaction) to break up the text and guide the reader through.*

   The Introduction now includes the following Section headers:

   1.1 Phase 4 of the Air Quality Model Evaluation International Initiative
   1.2 Deposition in the Athabasca Oil Sands Region – Evidence for pH-modulation
   1.3 Foliage Surface pH and Alkaline Dust
   1.4 Particle and Gas Deposition and Foliage
   1.5 Removal of Deposited Material from Foliage
   1.6 Foliage Surface Water
   1.7 Overview of the Current Analysis

Section 2.2

1. *Are the observations from Hayden et al., (2021) and Gordon et al., (2023) those that are used in Figure 9 (Oski-otin, YAJP and DWEF)? It would be useful to include these sites on Figure 1 along with a short description of the site characteristics in the text.*

   The revised site locations have been included into the leftmost panel of the revised Figure 9 for reference. Note that the original manuscript Figure 9 has become Figure 10 in the revised manuscript.

Section 3 – Dry deposition algorithms

1. *L299: $Hsp*$ ≈ 105 M atm^{-1}.* The given text has been changed to "To obtain H^*_{sp} for SO_2 ($H^*_{SO_2}$, where $H^*_{SO_2 neutral} = H^*_{SO_2 n} \approx 10^5$ M atm^{-1}) "

Here, is "M" mol L^{-1}?

1. *L340: "To obtain $HSO2n* = HSO2 * n$ (= 105 M atm-1 340) as…" Should this be "To obtain $HSO2* = HSO2 * n$ (= 105 M atm-1 340) as…" ?* See above comment. The line 340 text has been changed to read "H^*_{sp} for SO_2" since revised line 299 makes the intent more clear and defines both $H^*_{SO_2}$ and $H^*_{SO_2 n}$.

Section 4 - simulating foliage pH

A short introductory sentence or two to describe the calculations necessary to simulate the foliage pH would be helpful here.

The following paragraph has been added to at the start of section 4:

"Several considerations must be addressed in order to carry out the calculation of foliage pH for the purposes of deposition velocity estimation, in addition to incorporating the dependence of V_d on $[H^+]$, as described in Sect. 3. Choices must be made regarding which foliage types should be subject to pH-dependent deposition. The accumulation of previously deposited material must be tracked across time steps (since this will determine the pH of the foliage surface in the current time step). This tracking must include both deposition fluxes to the foliage and the removal of deposited material via precipitation (washoff). The amount of liquid water adhering to leaf surfaces must also be estimated, from both meteorological and thermodynamic sources. The pH calculation must take into account high-concentration thermodynamic chemistry, since the water amounts can be sufficiently small that non-ideal solution chemistry solvers must be employed. The formula representing deposition must make use of the resulting pH values, and account for varying types of foliage within each model grid cell; the extent to which deposition specifically to the foliage portion of the surface is resolved may depend on the algorithm (for example, the gas and particle deposition algorithms may differ in this respect). These aspects of the simulation of foliage pH are discussed in Sections 4.1 through 4.5. Note that none of the pH calculations are employed in the Base Case simulations, which use the default pH value of 6.68."

1. *L595: "below" Suggest pointing to the specific section/subsection.*

   Changed to "below (Sect. 5.1).

Section 5.1

1. *L733-737: Essentially repeats Figure 5 caption.*

   Submitted Figure 5 has been moved to the Supplement (revised Supplement Figure S2), and is mentioned only briefly in the revised main document.

2. *Figure 5: Masking out the ocean would help the reader interpret Figures a, b, d and e (which is mislabelled 'd')*

   The revised Figure 5 (now Figure S2) includes the masking as suggested by the reviewer, and the panel labelling has been corrected.

3. *L786-789 and L798-801: This information should just be in the Figure 6 caption.*

   The given text was removed from the manuscript, and the revised manuscript Figure 5 caption now reads, "**Figure 5: (a):** The deposition velocity of SO_2 ($\boldsymbol{V_{d_{SO2}}}$, cm s^{-1}) as a function of foliage (leaf) pH; data are from May 2018. $\boldsymbol{V_{d_{SO2}}}$ has been separated into 8 discrete bins, and the corresponding box plots give the range of leaf pH (lower x-axis) within each bin. The whiskers, box boundaries, central vertical line, etc. from left to right for each box are the 5th percentile, 25th percentile, median, 75th percentile, and 95th percentile, respectively. This format for box plots is applied henceforth. Each box is superimposed over a shaded bar that gives the percentage of $\boldsymbol{V_{d_{SO2}}}$ in the corresponding bin (upper x-axis). **(b):** The foliage (leaf) pH as a function of the estimated leaf water layer thickness ($\boldsymbol{L_w}$, **m**m). $\boldsymbol{L_w}$ has been separated into 12 discrete bins, and the corresponding box plot gives the range of leaf pH in the corresponding bin. The shaded bars give the percentage of $\boldsymbol{L_w}$ in each bin (right y-axis)."

4. *Figure 6: Could (b) and (d) be oriented the same way as (a) and (c)? Also, the text on the shaded bars is very difficult to read. It would also be helpful if (b) and (d) used the same scale for the right y axis.*

   The panels in revised manuscript Figure 5 have been rotated so that pH is on the x-axis for all panels, and the scales in (b) and (d) have been matched, as per the Reviewer's suggestion. The text within the shaded bars in panels (a) and (c) was removed in the revised Figure – the numbers are a bit redundant in that the same information is given by the length of the grey bars, and the smaller numbers are not well represented in a real number format.

5. *L825-829 – Point to the figures that are being discussed.*

   The lines in the submitted manuscript included "see Sect. 5.3" towards the end of this passage, to let the reader know that the details (and Figures) supporting the statement follow in that section. We've moved the "see Sect. 5.3" to the end of the first sentence in this paragraph, to make this more clear.

Section 5.2

1. *L857-861: Information should just be in the Figure 8 caption.*

   Submitted manuscript Figures 7,8,10,13 were combined into revised manuscript Figures 6, 7, 8, and the text accompanying them was extensively modified (see response to General Comment 4, above).

2. *L875-880: If the statement beginning "As the pH increases above 9.0…." is referencing Figure 2, please cite Figure 2 in the text.*

Text changed to "As the pH increases above 9.0, both $r_{cut}(\mathrm{SO}_2)$ and the canopy resistance $r_c(\mathrm{SO}_2)$ approach zero (see Figure 2). ".

3. *L889-893: Should only be included in the Figure 9 caption.*

   The main text now reads: "Three locations in the 2.5 km model domain; Oski-otin (near the village of Fort Mackay), the York Athabasca JackPine site (YAJP), and a downwind evergreen forest location near the Saskatchewan border (DWEF) are superimposed on the monthly average pH field in Figure 8(a), and their corresponding diurnal cycles of $V_{d_{\mathrm{SO}_2}}$ for the months of May and June are shown in Figure 9.", and the Figure 9 caption reads, "The diurnal cycle of model-predicted $\boldsymbol{V_{d_{\mathbf{SO_2}}}}$ at (a, d) Oski-otin, (b, e) the York Athabasca Jack Pine (YAJP) Tower, (c, f) a downwind evergreen forest (DWEF). The first row (a, b, c) shows the predicted $\boldsymbol{V_{d_{\mathbf{SO_2}}}}$ for May 2018 while the second row (d, e, f) shows June 2018. The box plot formatting in Figure **9** is consistent with Fig. **5** and solid lines indicate the mean $\boldsymbol{V_{d_{\mathbf{SO_2}}}}$ in each hourly bin.

4. *L889-921: I think this text should be split into a separate subsection. Something like "5.X Comparison of modelled vdSO2 with surface monitoring network observations"* The section was completely rewritten in response to the Reviewer's comment regarding amalgamating submitted manuscript Figures 7, 8, 10 and 13, and the subsections of section 5 were reordered to allow better flow of the material. We hope this addresses the Reviewer's concerns.

Section 5.3

Figures 11 and 12 show that there are only small changes in SO2 concentrations between the base and CALCCO3_Lw_high simulations at most of the sites and only five sites where CALCCO3_Lw_high substantially reduces bias against the observations.

We respectfully disagree with this assessment, though it may depend on what threshold is set for "substantially reduce". The tables incorporated into the right-hand side of the panels of submitted manuscript Figure 12(b,d) show reductions in the magnitude of the SO2 bias as 13 and 15 out of 21 stations, respectively. We also note that the general impact of CALCCO3_Lw_high was that of decreases in SO2 concentrations at *all* sites (that is all biases are shifted towards lower, or more negative numbers; see revised Figure 11 tables). In some cases the bias was already negative, and hence the bias became worse (black tabulated numbers, revised Figure 12 tables), while cases where the bias was positive without the pH correction became less positive or small negative (red tabulated numbers, Figure 11 tables). The following sentence was added to submitted manuscript

Figure 12 (now revised manuscript Figure 11): "A general result of the comparison is that NMB values "shift towards negative numbers" – that is, all NMB values are decreasing - for stations where the base case NMB was positive this usually results in a smaller magnitude positive or negative NMB (red tabulated numbers), and for stations where the base case NMB was negative, the NMB becomes more negative (black tabulated numbers)." We also note that the revised text corresponding to the revised Figures 6, 7, 8 (from the amalgamation of submitted manuscript Figures 7, 8, 10 and 13) notes that the changes in SO2 concentration and SO2 deposition flux are more than a factor of 2 throughout the model domain.

This is despite the better agreement between CALCCO3_Lw_high and observed VdSO2 shown in Figure 9. Is there anything linking the five WBEA sites where CALCCO3_Lw_high reduces bias against the observations and Oski-otin, YAJP and DWEF? E.g. location or vegetation type?

We note that submitted and revised manuscript Figure 9 compares CALCCO3_Lw_high and Base Case model predictions, not CALCCO3_Lw_high with observed VdSO2. We have revised the Figure caption to make this clear ("The diurnal cycle of model-predicted $\boldsymbol{V_{d_{SO_2}}}$ …". We've plotted these three locations on the new Figure 8 of the revised manuscript (since this Figure appears before Figure 9). The main point from Figure 9 is that the impact of surface pH decreases as the calculated pH value decreases (approaches neutrality or more acidic conditions) – this is more clearly seen by comparing the three locations as given in revised Figure 8 to the corresponding time series of revised Figure 9: Oski-Otin and YAJP are located in the region with the highest predicted pH (~9.0), while DWEF, further downwind and further away from the sources of fugitive dust, shows a much smaller impact on $\boldsymbol{V_{d_{SO_2}}}$. The text referencing Figure 9 now begins "Three locations in the 2.5 km model domain; Oski-otin (near the village of Fort Mackay), the York Athabasca JackPine site (YAJP), and a downwind evergreen forest location near the Saskatchewan border (DWEF) are superimposed on the monthly average pH field in Figure 8(a), and their corresponding diurnal cycles of $V_{d_{SO_2}}$ for the months of May and June are shown in Figure 9. The DWEF site was selected to represent a downwind location where anthropogenic dust deposition is minimal and foliage water layers are often acidic to near neutral. This contrasts with both YAJP and Oski-otin, where deposition of alkaline dust frequently raises the foliage pH above 7.5 (see Fig. 6-8(a))."

I also found Figure 12 hard to interpret due to the small font and symbol sizes. Perhaps the authors could consider i) including just Figure 12b (the results from June that are also presented in Figure 11) and ii) summarizing the results from Figures 12a, c and d?

We have modified the submitted manuscript Figure 12 (which appears as Figure 11 in the new manuscript) to show the June and August values (b,d), and have included the May and July figures as Figure S10 in the revised Supplement.

Can the authors suggest a reason/s why CALCCO3_Lw_high reduces bias against the atmospheric SO2 observations at some sites and months, but not others? Are these sites linked by location or vegetation type? Further, is there anything linking the five WBEA sites where CALCCO3_Lw_high reduces bias against the observations and Oski-otin, YAJP and DWEF? Again, location or vegetation type?

As noted above, the overall effect of the pH correction is to result in a *shift* in bias from positive towards more negative numbers, at all stations. Stations which had a base case positive bias thus tended to have a CALCCO3_Lw_high smaller magnitude positive or negative bias, and stations with a base case negative bias saw this negative bias increase in magnitude with the use of the correction. All sites in this respect have a similar response in the "direction" of the bias. There are therefore other factors aside from the pH correction underlying why the initial base case bias was positive or negative. We have added the sentences, "A general result of the comparison is that NMB values "shift towards negative numbers" – that is, all NMB values are decreasing. For stations where the base case NMB was positive this usually results in a smaller magnitude positive or negative NMB (red tabulated numbers), and for stations where the base case NMB was negative, the NMB becomes more negative (black tabulated numbers). Additional factors aside from the pH effect studied here are therefore likely to result in the remaining station-to-station variations in NMB."

1. *L942-943: Should only be included in the Figure 11 caption*. The sentences in the text now read, " Time series of both observed and simulated mean diurnal cycles of the surface SO_2 concentration at 20 different WBEA continuous monitoring stations (Figures 10, S7, S8, S9, and 11, S10) show that the pH-modulation effect has the most pronounced impact on the mean surface SO_2 concentrations (relative to the Base Case) at the stations BGFM, FMCS, HRZN, MUSK, FTHL and WAPI, though improvements in NMB can be seen in 13 and 15 out of 21 stations (Figure 11(a,b)). At the six stations with larger improvements, CALCCO3_Lw_high yields a substantial reduction in the normalized mean bias (NMB) relative to the Base Case (Table 3)." The caption for the Figure now includes the sentence, "Identical plots for May, July and August 2018 are provided in the supplemental material (Figures S5, S6, S7, respectively)."
2. *L947-949: Please consider tabulating these results*. Done. This appears as Table 3 in the revised document.

3. *Are the results from Hayden et al., 2021 comparable with the results presented here?*

   Yes, they are. As stated in both the original and revised manuscripts, "This enhancement is consistent with aircraft-based measurements reported by Hayden et al., (2021), who found that an earlier version of GEM-MACH, lacking the pH $V_{d_{SO_2}}$ enhancement, underpredicted the dry deposition flux of total oxidized sulphur (TOS) by 2 to 14 times".

4. *L972:979: Should only be included in the Figure 12 caption*.

   Most of the information flagged by the Reviewer has been moved to the submitted manuscript Figure 12 (revised manuscript Figure 11) caption as requested. The first two sentences, "Figure 11 presents Taylor diagrams for (a) July and (b) Aug 2018 (see Supplement Figure S10 for May and June); these summarize graphically how closely modelled surface SO_2 concentrations match observations (Taylor, 1991). The diagrams simultaneously depict three key statistics: the correlation coefficient, the centred root-mean square difference (CRMS), and the standard deviation (amplitude of variation).," were retained for the benefit of readers unfamiliar with Taylor diagrams. The revised Figure 11 caption now reads, "Figure 11: Taylor Diagram of SO_2 for (a) June 2018, (b) Aug 2018 (see Figure S10 for May and July). The equations used to compute these metrics are provided in Table S3. In each panel, the "star" symbol on x-axis marks the reference point for a perfect model, corresponding to a correlation = 1.0, a CRMS = 1.0 and a normalized standard deviation = 1.0. Arrows illustrate changes in model performance from the Base Case (arrow start) to CALCCO3_Lw_high (arrow end). A trajectory that moves closer to the reference point indicates improved agreement with observations (red arrows). The CRMS is proportional to the distance from the star; concentric circles centred on the star represent contours of constant CRMS. The standard deviation is measured radially from the origin. The different WBEA stations are denoted by different symbols, with each arrow pointing from the Base Case to CALCCO3_Lw_high locations of station performance on the diagram. Red (black) arrows denote an improvement (deterioration or no change) in the normalized mean bias error (NMB) for CALCCO3_Lw_high compared to the Base Case. The value of the NMB for each station is shown in the legend after the station name as (Base Case → CALCCO3_Lw_high), with red text likewise representing improvements in the NMB when using CALCCO3_Lw_high compared to the Base Case. The azimuthal distance represents the correlation (i.e., r) starting from 0.0 and increasing clockwise to 1.0. The radial distance from the origin gives the normalized standard deviation of the

model at the station. Each station standard deviation has been normalized by the observed standard deviation at that station. Thus, the observed standard deviation is always at (1.0, 1.0) represented by the star on each plot. May and July Taylor diagrams are given in the Supplement, Figure S10."

5. *L991-1002: Could the model's wet deposition parameterization also be a source of uncertainty?*
   We agree with the reviewer that this should be mentioned – we did have a mention of part of the wet deposition algorithm in our point iii, but not the cloud to rain conversion rate. We've modified point iii to read: "chemical transformation of SO_2, particularly the aqueous-phase oxidation to sulphate in cloud droplets or within emission plumes, along with the cloud to rain conversion process governing wet deposition".

Reviewer 2:

General comments

The manuscript entitled "Alkaline dust deposition to foliage surfaces likely enhances the dry deposition velocity of SO_2: An investigation in the Alberta Oil-Sands Region using the GEM-MACH air-quality model" by Stefan Miller et al. investigates the effect of alkaline dust deposition on SO_2 dry deposition velocity using the GEM-MACH model. This detailed examination in the AOSR contributes to our understanding of the role of deposition processes, and I am generally inclined to recommend publication of this manuscript. I would, however, encourage the authors to address the following specific points, particularly those related to presentation quality.

Major comment

- *With respect to the analysis of deposition, the deposition flux (Fig. 13) is an important quantity, and I fully agree with the authors' choice to emphasize this in their analysis. However, a limitation of the current approach is that the flux is presented as a mean field over the study period (i.e., monthly averages in this manuscript). To better capture the variability of deposition, I suggest also examining the total deposition amount accumulated over the target period (e.g., monthly totals). This perspective may reveal different characteristics compared to the averaged fields and could highlight additional impacts in the sensitivity experiments.*

We agree that total monthly deposition rather than monthly average deposition is a more appropriate quantity for describing the magnitude of deposition flux impacts of the work. We have consequently changed the deposition flux scales on the revised version of these Figures to units of equivalents per hectare per month (equivalents are charge units, where each mole of deposited S contributes two moles of equivalent charge). This does not result in large differences in the resulting panel images as the Reviewer suggested might be the case, since this amounts to multiplying the original displayed field by a constant factor (2 x 1E4 x number of hours in the month). That is, the monthly average hourly deposition flux field is the sum of the individual hourly fluxes divided by the number of hours in the month, while the monthly accumulated deposition flux field lacks the division in the numerator. Converting to total accumulated deposition thus scales the numbers, but does not change the comparison significantly (we note that the difference in the number of days in June (30) versus the other two months (31) results in a 3% change in the colour bar scales between these months, but this is not a significant difference.

Note that the revised fields appear in the Revised Supplement Figures S3, S4, S5, in response to comments from Reviewer 1.

Specific comments

- *Line 269: To avoid confusion, it would be helpful to note explicitly that rac is discussed later and is not introduced here (see eqs. (3)–(8))*.
  The sentence before equation (3) now reads: "The formulae for these terms (aside from r_{ac}, defined in equation (10)) are, respectively (Makar et al., 2018; Clifton et al., 2023)*"*. Note that the sentence now appears in Supplement section S1.0 in response to requests from the other Reviewer to reduce the size of the main text.

- *3: The colored corners (light blue and light green) in each panel are unclear. Please explain what these colors represent in the figure or caption*.

  Fair point. The boundary regions were areas of "no data" for the land use field being depicted. The graphics system employed in the submitted Figure 3 used a default colour of green for land and blue for water, wherever the model field is not present. That plus the use of green shades for the model fields centered in each panel creates confusion, so in the revised version of Figure 3, the individual panels have been "clipped" to only show the area with the valid model input fields on the 2.5km grid on which the model operates, and none of the surrounding region.

- *Line 605: Please clarify how the minimum value of the multiplication coefficient of 0.5 by TK was determined*.

  The values of TS and TK are in molar units, and the salt which they may form (K2SO4) has half as much sulphate as potassium, by moles. Consequently, the maximum amount of potassium sulphate which may form is the minimum of the sulphate amount and half of the potassium amount. This information has been added as a footnote to the revised text, "The values of TS and TK are in molar units, and the salt which they may form (K_2SO_4) contains half as many moles of sulphate as potassium. Consequently, the maximum amount of potassium sulphate which may form is the minimum of the sulphate amount and half of the potassium amount."

- *Lines 643–654: Please clarify the actual calculation method used for the charge balance. Is HETP applied in the "Base Case" simulation? This section feels somewhat long; it may be helpful to summarize the key points in a table (e.g., revising or expanding Table 2) for clarity.*

  Section 4 was extensively re-written in response to suggestions from the other Reviewer to streamline the methodology discussion (see responses to Reviewer 1, above). Both the charge_adj and Lw_low tests no longer appear in the main text, and instead the results of the tests are summarized, with the focus now being on the CALCCO3_Lw_high simulations throughout the main manuscript. In the revised manuscript, we have made it clear that the Base Case simulation uses no pH adjustment (HETP is not applied to the problem of foliage surface pH calculations), "Note that none of the pH calculations are employed in the Base Case simulations, which use the default pH value of 6.68.".

- *5: It would be helpful to label the top and bottom panels explicitly as "May 2018" and "June 2018," respectively, because the two panels appear very similar at first glance.* Done – labels added as suggested. A mask has also been applied to the water bodies to allow the reader to focus on the land surfaces (suggested by Reviewer 1). Note that submitted manuscript Figure 5 has been moved to the revised manuscript Supplement Figure S2, in response to a request from the other Reviewer.

- *Lines 759–779: The discussion of "charge_adj_Lw_low" (#3 in Table 2) and "charge_adj_Lw_high" (#4 in Table 2) is somewhat difficult to follow. Consider adding a figure analogous to Fig. 5 to illustrate these cases more clearly.*

  In response to a comment by the other Reviewer, the details of the charge_adj_Lw_high, charge_adj_Lw_low and CALCCO3_Lw_low simulations were

summarized briefly in the revised text in favour of focussing the revised manuscript on the recommended configuration, CALCCL3_Lw_high. Sections 4 and 5 were extensively reworked to improve readability, with a reorganization of the order of presentation of material in Section 4, and a reduction in the size of Section 5.1 to focus only on the CALCCO3_Lw_high results. The intent of the text in that line range was to point out that our tests showed that the charge_adj methodology generates unrealistic pH values and hence should not be used.

- *6: Please unify the x-axis range for "Leaf pH" so that (a) can be directly compared with (b), and (c) with (d).*

  The original panels did have exactly the same ranges for Leaf pH, but panels (a) and (c) had these on the x-axis, while panels (b) and (d) had the leaf pH on the y-axis. In the revised Figure (Figure 5 in the revised manuscript), we've shown only the CALCCO3_Lw_high results (due to the other Reviewer's request to simplify this section and focus on the recommended configuration), and we've swapped the axes on the Lw panel (revised manuscript Fig. 5(b)) so that Leaf pH appears on the x-axis similar to Fig. 5(a), with the same (identical) pH ranges as before, on all panels.

- *Line 947: The discussion based on NMB can also be inferred from Fig. 12. I therefore recommend explicitly citing Fig. 12 together with Fig. 11 here. This cross-reference will be helpful for readers.*

  A good point – we've summarized the NMB changes as a new Table (3), and added to the caption "(see also Fig. 11(a,b))" to alert the reader that the same numbers are shown also in this Figure. Note that the Figure numbers were updated in response to a request from the other reviewer to amalgamate the panels of submitted manuscript Figures 7, 8, 10, and 13.