

Author responses to reviewer comments on “Decreasing seasonal cycle amplitude of methane in the northern high latitudes being driven by lower latitude changes in emissions and transport” by Dowd et al.

We would like to thank the reviewers for their time and feedback to help improve our manuscript. We have responded to both general and specific comments in the tables below. Changes to the text in the manuscript are highlighted in blue and the line numbers refer to the updated text. Please note that Latexdiff has not highlighted the changes to the bibliography which are described in the responses below.

Reviewer 1’s Comments:

General Comments

“The manuscript by Dowd et al., entitled “Decreasing seasonal cycle amplitude of methane in the northern high latitudes being driven by lower latitude changes in emissions and transport,” presents an interesting new study that analyzes changes in the seasonal cycle of methane concentration. The authors use an atmospheric chemistry-transport model with an inverse model to explore how and why the seasonal cycle amplitude (SCA) in methane concentration has changed from 1995-2020. The topic is of interest to scientists from a wide range of disciplines and backgrounds in the CH₄ community. The model description is detailed and clear, but I have a few questions about the results and conclusions.

We would like to thank Reviewer 1 for their positive feedback and useful comments. We have addressed general comments in the table below and think that these revisions have clarified and improved our manuscript.

	Comments	Response
1	The manuscript's structure is not well-balanced, with the first half discussing global patterns of SCA and the second half focusing only on changes in the 60N-90N region. The lack of in-depth discussion or explanation for why the region below 60N is not interpreted is also reflected in the abstract, which suggests a global increase in SCA by 2.5 ppb without further explanation.	<p>Thank you for highlighting the imbalance in our manuscript. We have tried to address this by highlighting throughout the text and the abstract that the change in the SCA in the northern high latitudes (NHL) is significantly different compared to the rest of the globe.</p> <p>We have added a line in the abstract that clarifies why an increase in the SCA is expected. See line 2:</p> <p>“The reaction between CH₄ and its main sink, OH, is dependent on the amount of CH₄ and OH in the atmosphere. The concentration of OH varies seasonally and due to the increasing burden of CH₄ in the atmosphere, it is expected that the SCA of CH₄ will increase due increased removal of CH₄ through reaction with OH in the atmosphere.”</p> <p>We also highlight in the abstract why the NHL is of interest, line 10:</p> <p>“TOMCAT reproduces the change in the SCA at observation sites across the globe. Therefore, we use it to attribute regions which are contributing to the changes in the NHL SCA, which shows an unexpected change in the SCA that differs from the rest of the world.”</p>

		<p>In paragraph 1 in Section 3.1 we explain that there are no regional or local patterns in ΔSCA in non-NHL compared to the NHL. We have explained why we expect an increase in SCA and that it is shown in the observations and highlight that the difference in behaviour in the NHL compared to the rest of globe is the focus of the study by adding the following text, line 119:</p> <p>“The reaction between CH₄ and OH is dependent on the amount of CH₄ available in the atmosphere. The combination of the increasing CH₄ burden in the atmosphere and the photochemically-driven seasonal variation of OH results in more CH₄ being removed from the atmosphere during the time of maximum OH. Therefore, an increase in the global mean SCA is expected due to the increasing atmospheric burden of CH₄. However, when we look at the ΔSCA latitudinally, there are large differences in the NHL compared to the rest of the world. The mean observed ΔSCA in the NHL was -4.0 ppb, which represents a 7.6% decrease between 1995 and 2020, and in the Non-NHL region the mean observed value of ΔSCA was 4 ppb, which is an increase of 11.5% for the study period. The reasons for this widespread contrasting behaviour in the NHL compared to the rest of the world is investigated in more detail in the forthcoming sections.”</p>
2	<p>The manuscript does not discuss how the model bias of the transport model affects interpretation. The evaluation shows that the BRW site is an outlier with a large overestimation of the decrease in SCA. With only four sites in the Northern High Latitudes (NHL), it is unclear how well the model captures the seasonal amplitude in the NHL and how model errors affect the results.</p>	<p>The model reproduces the decrease in the SCA within the uncertainty when compared with observations, so we are confident that the model transports the emissions with sufficient accuracy. We have addressed this by adding this to the results section, Section 3.2 line 275:</p> <p>“Despite some differences between the model and observations in the NHL and Non-NHL regions, the model still captures the change in the SCA across the globe, almost all within 1σ uncertainty of the observations. We are confident that the transport in the model is sufficiently accurate to inform our conclusions.”</p> <p>We also agree that ΔSCA at BRW is an outlier, however it is within the 1σ uncertainty of observations (TOMCAT: -19.0 +/- 9.9 ppb and Observations: -6.8 +/- 5.5 ppb). Our original results show that Canada, the Middle East and Europe are the largest contributors to the decrease in the SCA in the NHL. When we remove BRW from the analysis it shows that Canada contributes to an increase in the SCA at the other NHL sites (ALT, ICE & ZEP). Europe and Middle East remain the largest contributors to the decrease in SCA. This implies that the proximity of the regions to the sites does have an effect on the final results. It also suggests that changes in Canadian emissions are having a larger effect on BRW compared to the other sites, which are being affected more by Europe and the Middle East.</p> <p>We are also interested in the NHL because it shows significantly different behaviour to the rest of the globe. The model also captures the different behaviour in the NHL shown by observations. We have added some text which discusses the impact of BRW on regional contributions to ΔSCA and why NHL is of interest in Section 4, line 410:</p> <p>“The TOMCAT tagged tracer simulations perform well when compared with observations (Fig. 4b). However, from Fig. 4b it is noted that BRW,</p>

		<p>which is situated in the NHL, is an outlier in the model, compared with other sites. The model does capture the change in the SCA within the observation uncertainties, but these are large for this site. To test the influence of BRW on our results we removed it from our analysis. We find that Canada is no longer the largest regional contributor to the decrease in the SCA in the NHL and, in fact, contributes to an increase in the SCA at the other sites (ALT, ICE & ZEP). However, Europe and the Middle East remain the largest contributors to the decrease in the SCA at ALT, ICE and ZEP (See Supplement Fig. S4). The removal of BRW from our analysis shows that local emissions are having the largest impact at this site. This is likely due to a strong decrease in emissions in DJF and an increase in emissions in JJA in Alaska and western Canada during the study period (See Supplement Fig. S6b). The seasonal changes in emissions over eastern Canada are different to Alaska and western Canada and it is likely that a different mechanism is having an effect on the other sites in the NHL. This test shows that the boundaries of the tagged tracer regions and the proximity of Canada and Europe to the NHL does have an impact on the results. For example, if Alaska was grouped into the North America (NAM) region, then NAM could be a large contributor to the decrease in the SCA due to the changes in emissions over Alaska. However, we include Alaska and Canada as one region due to their similar biomes and meteorology. Despite some differences between the model and observations (e.g. at ALT and BRW), TOMCAT does capture the significantly different behaviour in the NHL compared to the rest of the globe. The change in SCA in the NHL is consistently lower compared to the rest of the globe, implying that increasing emissions, both local and non-local, are impacting the NHL differently.</p> <p>We have also added the following text in the discussion to reflect the effect of removing BRW on the largest contributors to the change in SCA in the NHL, Section 4, line 447:</p> <p>“Canada has the largest negative contribution to the ΔSCA NHL due to emissions (-2.97 ppb), however we have shown that this region predominantly affects BRW.”</p>
3	<p>The manuscript does not discuss the role of Russia in the change in SCA, despite having the largest wetland emissions in the northern high latitudes, high oil and gas emissions, and severe biomass burning events in Siberia. Is that partly because there are no in-situ sites for that region so you underestimate the regional contributions from Russia with the taggers?</p>	<p>In our analysis we find that changes in emissions from Russia are not the cause of the observed decrease in the SCA at NHL sites, see Figure 5b. However, changes in transport from Russia do contribute to a small decrease in the SCA, with a ΔSCA of -0.6 ppb (Table A1).</p> <p>In the inversion, 80 surface sites were used to constrain the model, which includes one site in Russia. This site is not used in our SCA analysis due to data only being available from 2011 onwards. The inversion and forward simulations also represent the transport of emissions well so the four sites in the NHL used in our analysis will be affected by Russian emissions.</p> <p>We have added some text in the discussion, line 501:</p> <p>“We find that Russia does not contribute to the decrease in the SCA in the NHL, despite it being a region that has large natural and anthropogenic</p>

		<p>emissions of CH₄. The Russian emissions used in the forward simulation are not locally constrained before 2011 but transport from Russia to the NHL sites is short (~2 weeks) because it is largely zonal (Jacob, 1999). The inversion and forward simulations represent transport of emissions well which means that the four sites in NHL will be impacted by Russian emissions throughout the study period, even when the inversion has few sites to constrain the model in this region. Our results show that changes in transport from Russia contribute to a small decrease in the SCA with a ΔSCA of -0.6 ppb (See Fig. 5b). This is a small contribution to the decrease in the SCA in the NHL, which is why we decided to focus on Canada, the Middle East and Europe as they have the largest contributions to decrease in the SCA in the NHL.”</p>
<p>4</p>	<p>The attribution of regional contribution is confusing, especially since more than 50% of the changes in SCA in the NHL come from unknown regions where the emissions originate. It is unclear how accurately the authors can rank regional contributions.</p>	<p>We chose to define CH₄ that takes a long time to reach the NHL to be “well-mixed” because the CH₄ has been transported for a long time and mixed thoroughly in the atmosphere. We suggest that after becoming well-mixed the CH₄ should no longer be attributed to a regional tagged tracer. We also used the background tracer to reduce the spin-up time in the model and tested the effects of decay rates into the background tracer in our sensitivity experiments. We have added some text to explain the reason for a background tracer and why we chose 9 months as the decay rate in Section 2.2, line 108:</p> <p>“Typical timescales for horizontal transport in the troposphere from the mid latitudes to the poles is approximately 1-2 months and interhemispheric transport takes approximately 1 year (Jacob, 1999). The 9-month decay rate was selected to maximise the opportunity for CH₄ to undergo long-range transport from emission locations to surface sites, whilst minimising the effect of well-mixed atmospheric CH₄ on the results. The background tracer allows us to reduce the spin-up time required in the model to reach steady state. Without the background allocation concentrations would continue to increase because it takes approximately 20 years for the CH₄ to reach steady state in the model.”</p> <p>We are also attributing the changes in the SCA from different regions by ranking recent contributions because after a certain time period, which we have chosen to be 9 months, the CH₄ has become well mixed and no longer ‘belongs’ to the region. We have added a line to Section 2.2, line 111:</p> <p>“The background tracer also allows us to regionally attribute recent contributions to changes in the SCA whilst accounting for well-mixed CH₄”</p>

Specific Comments:

	Comments	Response
1	Abstract: In the abstract, it would be helpful to add an explanation of why higher methane concentration leads to a corresponding increase in seasonal amplitude.	This has been addressed from Reviewer 1's General Comment 1. See line 2 in the abstract. "The reaction between CH ₄ and its main sink, OH, is dependent on the amount of CH ₄ and OH in the atmosphere. The concentration of OH varies seasonally and due to the increasing burden of CH ₄ in the atmosphere, it is expected that the SCA of CH ₄ will increase due increased removal of CH ₄ through reaction with OH in the atmosphere."
2	Abstract: The total contribution in the abstract is not 100%, and the regions or processes missing need to be specified.	We have added this line to the abstract, line 15: "The remaining contributions are due to changes in emissions and transport from other regions."
3	Line 245-248: Regarding initialization, it would be beneficial to remove the beginning period to exclude the initialization effect	The initialisation period of the model (1983-1994) is not included in the ΔSCA analysis. This explained line 116.
4	Paragraph starting Line 274: The paragraph over results (Line 280) is confusing. Please rewrite it to be more clear about whether positive means the emissions lead to an increase in DeltaSCA or a decrease in DeltaSCA.	We have edited the wording about positive and negative contributions from the different TOMCAT simulations, line 306: "Changes in the transport of emissions from North America and Russia have also contributed to the decrease in the SCA between 1995-2020 in the NHL, however the changes in emissions from these regions contribute to an increase in the SCA. The change in SCA due to emissions is larger in magnitude than the contribution from transport, resulting in overall increase in the SCA in the NHL from these regions. The TOM_transport contribution to ΔSCA in NHL from Canada and Europe is 0.24 ppb and 0.77 ppb, respectively, resulting in an increase in the SCA in the NHL due to changes in transport. However, changes in emissions result in an overall decrease in the SCA from these regions. This is due to the magnitude of the decrease in SCA being larger than the contribution from transport."
5	Line 284: There is a typo in Line 284.	This has been corrected.
6	Line 286: It would be beneficial to have a conclusion sentence at the beginning of Line 286.	We have added a summary line at the beginning of Line 316: "The TOMCAT simulations (TOM_regional and TOM_transport) show the largest contributions to the decrease in ΔSCA in the NHL are mostly due to changes in emissions from Canada, the Middle East and Europe."
7	Line 264: Section 3.4 title needs to clarify whether it discusses DeltaSCA for the NHL.	We have changed Section 3.4's title to: "Regional Contribution to ΔSCA in Northern High Latitudes"
8	Figure 5: If Figure 5 is for the NHL, it needs to be described clearly in the figure caption.	We have edited the caption for Figure 5: "The contribution of the (a) background tracer and (b) regional tagged tracers to CH ₄ Δ SCA (ppb) for 1995-2020 as a mean across all sites in the latitude band. The blue bars show the NHL and the orange bars are the two Non-NHL latitude bands in the northern hemisphere. The hatched bars show the contribution from transport (TOM_transport) and the solid

		colour represents the contribution from emissions (TOM_regional). Note, (a) and (b) have different scales.”
9	Line 398: ‘Emissions’ or ‘anthropogenic emissions’?	Thank you for noticing this, it should be anthropogenic emissions, we have changed the citation to the original study (Lu et al. 2021) on line 458: “Lu et al. report that top-down estimates from satellite data show a decreasing trend in anthropogenic emissions for 2010-2017.”

Reviewer 2’s Comments:

General Comments

“Dowd et al first use NOAA measurements of methane (CH₄) concentration to quantify the changes in the observed seasonal cycle amplitude (SCA) at available surface sites throughout the globe over the past ~25 years. The northern high latitude (NHL) sites show a large reduction in SCA over this time, while most other sites show a range of increases. They then compare the observed changes with those from a chemical transport model driven by optimized CH₄ fluxes. Various model set-ups are used to evaluate the skill of the model and determine the causes of the negative dSCA at the NHL sites. The authors find that the NHL sites are mostly impacted by transported well-mixed background and more recent emissions from Canada, Europe, and the Middle East. The study highlights how impacts on the NHL can be used to isolate both local and non-local CH₄ emissions changes.

Overall, the paper contains content and is of a quality and significance appropriate for publication in ACP. Both the observational and extensive modeling components are well done. My main concerns relate to the description of the results and inclusion of appropriate caveats within the discussion. Streamlining certain sections and providing clarifications (in text and in figure captions) will improve the readability of the paper and increase its impact for the reader.”

We would like to thank Reviewer 2 for their positive and constructive feedback. We have condensed and refined some paragraphs in our results sections. We have also developed our discussion through Reviewer 1 and Reviewer 2 comments, which now includes the appropriate caveats for the study, particularly surrounding the proximity of the regions to the observation sites. Please see below our responses for the specific comments.

Specific Comments:

	Comments	Response
1	Line 16: It is not obvious from the abstract alone why the negative change in SCA is counter-intuitive (oxidation of increasing CH ₄ concentration leading to increasing SCA is only mentioned later).	We have added a line in the abstract that clarifies why an increase in the SCA is expected. See line 2: “The reaction between CH ₄ and its main sink, OH, is dependent on the amount of CH ₄ and OH in the atmosphere. The concentration of OH varies seasonally and due to the increasing burden of CH ₄ in the atmosphere, it is expected that the SCA of CH ₄ will increase due increased removal of CH ₄ through reaction with OH in the atmosphere.”
2	Line 42: “the observed CH ₄ seasonal cycle” Do the authors mean cycle of concentrations or flux/emissions? Check to make sure this distinction is clear throughout the paper. If “CH ₄ ”	Yes, we agree this is unclear. Throughout the text we are referring to concentrations when we use “CH ₄ ” alone, and we have added text at line 56 to make this clear: “Note that throughout this text we are referring to concentrations when we use “CH ₄ ” alone.”

	alone implies “CH4 concentration”, state this early on.	
3	Line 63 & Figure 2: I was confused to see 80 sites mentioned here after only 22 were depicted in Fig 2 (directed to in line 60). The reason for this difference is mentioned later but expanding the Fig 2 caption to state that only the sites shown were used to calculate the SCA would reduce initial confusion.	<p>We have edited Figure 2’s caption:</p> <p>“A map showing the 18 different regions selected for the tagged tracers, 22 NOAA surface observation site locations (blue) and the independent observations site locations (red). The observation sites shown are those used to calculate SCA from 1995-2020.”</p>
4	Line 79: How does the choice of the specific regions and their boundaries impact the results?	<p>It is difficult to quantify the impact of region boundaries on the results. However, when we tested the impact of BRW and MHD on our results it shows that proximity to the surface sites and the boundaries of specific regions do have an impact on the final results. See response in Reviewer 1 General Comment 2 and Reviewer 2, Comment 11.</p> <p>The justification of the selected regions is described in Section 2.2, paragraph 1. We have shown that Canada and Europe have an impact on the regional contribution to ΔSCA in the NHL due to their proximity to the observation sites, when we tested the impact of BRW and MHD on our results (see Reviewer 1’s General Comment and Reviewer 2 Comment 11). However, the Middle East is the 2nd largest contributor to a decrease in the SCA, in contrast to Russia, so proximity is not necessarily the main driver of the results.</p> <p>The regions do vary in size considerably, particularly in countries and continents that have been split up into smaller regions due to their emission type. We have normalised the ΔSCA contribution by area to assess the impact this has on the results. We have added a figure showing the normalised regional contributions to the site in the latitude bands to the supplementary material (Fig. S3). We have also added a few sentences to explain this result in Section 3.4, line 313:</p> <p>“We also assess the effect the size of the regional tracers has on our results by normalising the regional contribution by area size. We find the largest contributors to the decreasing SCA in the NHL are still due to changes in emissions from Canada, the Middle East and Europe (see Supplement Fig. S3).</p>
5	Line 142: Are the results impacted by running the inverse model over each calendar year when the seasonal cycle in the southern hemisphere spans two calendar years? or is more important only that the inversion is run consistently over each year?	<p>The results are not affected by running the inverse model over each calendar year because the inverse model results are consistent in terms of CH₄ mass across time. There are also spin-up and spin-down months included in each year’s inversion to give the transport of fluxes time to reach the measurement sites. We have edited the final lines in Section 2.3 line 154:</p> <p>“Each inversion overlapped with the following one by 2 months to give the transport of fluxes time to reach measurement sites. The overlapping months were initialised using 3-D fields provided from the correct date in the previous year so the total CH₄ burden was conserved across each year.”</p>

6	Line 154: Is this the top-down or bottom-up value from Saunois et al., 2020?	This is the top-down estimate. We have clarified this in the text, Section 2.3 line 164: “The wetland fluxes were then masked to remove emissions which overlap with rice emissions and then scaled back up to 180 Tg to match the top-down mean value from the Global Methane Budget (Saunois et al., 2020)”
7	Line 205: “the SCA is increasing globally” should more precisely be “the global mean SCA at available sites is increasing”.	Changed to Reviewer 2’s suggestion, line 216: “We find that the global mean SCA at available sites is increasing but there are different regional trends, for example in the NHL the observed Δ SCA decreased at all sites between 1995-2020 (Fig. 3)”
8	Figure 4: Site labels should be manually adjusted to avoid overplotting. Expand caption to clarify that 4a shows the mean SCA and interannual variability at each site over a certain range of years.	Figure 4 has been adjusted so that site labels do not overlap. The caption has also been expanded to: “Comparison between simulated and observed (a) CH ₄ SCA (ppb) and (b) CH ₄ Δ SCA (ppb). The SCA shown is the mean SCA between 1995-2020 and Δ SCA is the change in SCA for the same period. The dashed black line represents the 1:1 line and the red line represents the least squares regression line. The error bars denote $\pm 1\sigma$, which represents the interannual variability between 1995 and 2020.”
9	Line 235: Do the NOAA sites perform better because they were also used to determine the optimized fluxes?	Yes, this is partly right. We have added some text, starting at line 271, to discuss this: “The model performs better at the NOAA sites partly because these sites are used to provide optimised fluxes in our model and because Δ SCA is calculated over a long time period of 25 years. The independent site at Mace Head (GC-MD) also performs well because Δ SCA was calculated over a period of 18 years. The independent sites in Siberia do not perform as well compared to the GC-MD and the NOAA sites because of the large variability in the SCA over the relatively short time period (6 years) of observations.”
10	Line 236: A bit of clarification is needed here. Only 2 of the 4 NHL sites match with the correct negative sign, but yes, they are all consistently on the relatively lower end - for both observed and model.	Thank you for this comment, we have edited the text to be clearer on line 260: “The model also captures Δ SCA well when compared with observations, including the decreasing Δ SCA and contrasting behaviour in the NHL shown by observations (Fig. 4b).” We are interested in the NHL because it is significantly different to the rest of the globe. We have tried to acknowledge this throughout the text and in Reviewer 1’s General Comment 1. We also find 3 out of 4 modelled sites in the NHL have a negative sign. See below the values of the modelled and observed Δ SCA: Model: BRW(-19.0 +/- 9.9 ppb), ICE (-4.0 +/- 2.8 ppb), ZEP (- 4.0 +/- 2.8 ppb) and ALT (1.7 +/- 2.6 ppb). Observations: BRW (-6.4 +/- 5.5 ppb), ICE (-0.1 +/- 3.7 ppb), ZEP (-5.4 +/- 3.7 ppb) and ALT (-4.4 +/- 3.2 ppb).
11	Line 248: Should this be four sites in the NHL? Or mention more	We were referring to the sites BRW and ALT. The sentence on line 266 has been edited:

	<p>specifically the two sites meant here? The ICE site is included with the NHL grouping but does not have a large negative observed dSCA. Why is this? Despite the NHL focus, ICE is not mentioned or discussed anywhere in the paper. Similarly, MHD (in upper northern mid-latitudes) is not discussed but does have a larger negative dSCA. Should MHD be grouped with the NHL sites?</p>	<p>“Despite the under- and over-estimations at these two sites (ALT and BRW) in the NHL, the mean value of ΔSCA in TOMCAT is -6.38 ppb in the NHL, which shows a larger negative trend in the SCA than the observed mean ΔSCA value of -4 ppb.”</p> <p>We have added in a sentence which discusses the smaller negative trend at ICE compared to the other NHL sites in Section 3.1, line 231:</p> <p>“All four sites in the NHL display contrasting behaviour and have a negative ΔSCA compared to the rest of the world; therefore, the NHL will be the main focus of our analysis.</p> <p>BRW, ALT and ZEP have a ΔSCA which ranges from -4 ppb to -5 ppb. The SCA amplitude at these sites are variable but have a strong decreasing trend. ICE has a smaller ΔSCA (-0.05 ppb) compared the other three sites in the NHL. There is a large decrease in the SCA during the first 4 years of the study and then the SCA value steadily fluctuates between ~30 and ~40 ppb. This results in no trend in the SCA for the rest of the study period leading resulting in a smaller negative ΔSCA compared to the other sites (See Supplement Fig. S2).”</p> <p>Whilst our focus remains on the region north of 60N the site at MHD does display a similar observed decrease in the SCA. We have included in the discussion the results when we add in MHD into our extended NHL (NHL_ext, 52N-90N), Section 4, line 427:</p> <p>“The main focus of our analysis was in the NHL, however observations Mace Head (MHD) also show a decreasing SCA, similar to what is observed in the NHL. When we included MHD in our analysis by extending the NHL (NHL_ext, 52N-60N), we found that its proximity to emission regions had an effect on the regional contribution to ΔSCA in the NHL_ext. Changes in emissions from Canada and the Middle East, and changes in transport from North America and the Middle East contribute the most to the decrease in the NHL_ext SCA. Europe contributes to an increase in the SCA in the NHL_ext (see Supplement Fig S5). This is because MHD is strongly influenced by local trends in emissions in western Europe (see Supplement Fig. 8b). The seasonal changes in eastern Europe are quite different to western Europe, which are likely to affect the sites north of 60N differently to MHD.”</p>
<p>12</p>	<p>Lines 236-252: In addition to addressing the questions directly above, this paragraph would benefit from being refocused on the main point, which seems to be the last sentence.</p>	<p>We have restructured this paragraph, starting line 260:</p> <p>“The model also captures ΔSCA well when compared with observations, including the decreasing ΔSCA and contrasting behaviour in the NHL shown by observations (Fig. 4b). As a result, we can use TOMCAT to inform us of what might be driving this significantly different behaviour in the NHL. There is a good correlation ($r=0.51$) between the model and observations; they almost always match within 1σ uncertainty of observations, with some outliers. At ALT, TOMCAT shows a ΔSCA of 1.7 ppb and this is due to the model underestimating the SCA when compared with observations, particularly at the beginning of the study period. At BRW the model has a</p>

		<p>much stronger negative ΔSCA when compared with the observations and this is due to the model overestimating the SCA at the beginning of the study period. Despite the under- and over-estimations at these two sites (ALT and BRW) in the NHL the mean value ΔSCA in TOMCAT is -6.38 ppb in the NHL, which shows a larger negative trend in the SCA than the observed mean ΔSCA value of -4 ppb. This is mostly due to the overestimation of the magnitude of the simulated ΔSCA at BRW. At WLG the model overestimates ΔSCA, again this is likely due to the model representation at this site. The time series of the SCA and its trend at each NOAA site can be found in the Supplement. The model performs better at the NOAA sites partly because these sites are used to provide optimised fluxes in our model and because ΔSCA was calculated over a long time period of 25 years. The independent site at Mace Head (GC-MD) also performs well because ΔSCA is calculated over a period of 18 years. The independent sites in Siberia do not perform as well compared to the GC-MD and the NOAA sites because of the large variability in the SCA over a relatively short time period (6 years) of observations. Despite some differences between the model and observations in the NHL and Non-NHL regions, the model still captures the change in the SCA across the globe, almost all within 1σ uncertainty of observations, and we are confident that the transport in the model is sufficiently accurate to inform our conclusions. Therefore, we will use TOMCAT to regionally attribute the changes in the SCA in the NHL.”</p>
13	<p>Section 3.3: Perhaps more explicitly state that this result forms the intuition (referenced in abstract and elsewhere) for what would be expected absent any transport or emissions changes.</p>	<p>In Section 3.3 we have added the line 289: “These results inform our expectation that the SCA is expected to increase with the increasing atmospheric burden of CH₄ due to more CH₄ being removed by OH.”</p>
14	<p>Figure 5a: Ideally, resize to be same scales as regions in 5b for a better comparison. Perhaps make 5a only 0.25 of the figure width to expand region bars. Also better differentiate that BKGRD is not a region – maybe remove “Region Code” from x-axis title.</p>	<p>We have adjusted the scales on Figure 5b but have not put it on the same scale as 5a because it is difficult to see the detail for the negative regional contributions, particularly for the TOM_transport simulations. We have also made the “Background” bar chart smaller and edited the x-axis title and corrected a typo in the legend.</p>
15	<p>Lines 296-306: The explanation of the offset between the concentration and emission seasonal cycle here is confusing and should be rewritten for clarity. Does a positive ISR always lead to a decreasing SCA? What about if transport is accounted for?</p>	<p>We have rewritten our explanation here. This effect between emissions and the SCA is most likely to occur for nearby regions such as NHL and NML. See line 328: “It is important to note that the emission seasonal cycle is out of phase with the concentration seasonal cycle at northern mid- and high latitudes, so a positive ISR in emissions leads to a decreasing SCA. This is because the CH₄ seasonal cycle minimum is during the summertime in the NHL, so increasing emissions during this time would raise the minimum value, thereby shrinking the seasonal cycle. Similarly, shrinking wintertime emissions would bring down the seasonal maximum, which occurs at the same time. This effect is mostly likely in regions near to sites in the NHL.”</p>

16	Figures 6-9: Including the yearly SCA values and trend for each region would help with interpreting the CH ₄ concentration and ISR changes already shown.	We have adjusted figures 6a-9a to include the mean SCA at sites across the NHL and its trend. We have also included the trend values for the tracer contributions, ISR and emissions in Figures 6-9.
17	Lines 307-335: Reorganize and condense the several regional paragraphs to focus on the main point – that emissions from these regions are decreasing the SCA in the NHL. Then describe how each region is different, what about the regional emissions is changing, and the corresponding uncertainties. What impact does the relative proximity of Canada and Europe to the NHL sites have on the results?	<p>We have reorganised and tried to refocus from your suggestions in Section 3.4 paragraph 4 onwards. Please see changes in the updated text, Section 3.4 lines 316–381.</p> <p>We have also addressed the impact of the proximity of regions to the sites in the NHL by removing BRW in one example and adding in MHD as another example. The results of these tests have been added to the discussion. See response in Reviewer 1 General Comment 2 and Reviewer 2 Comment 11.</p>
18	Section 3.5: Which of these sensitivity experiments is more realistic? Are there any examples from the literature that 9 months is an appropriate choice?	<p>The typical horizontal transport times in the troposphere from the midlatitudes to the poles is 1-2 months, whereas interhemispheric transport is approximately 1 year (Jacob, 1999). We chose a 9 month decay rate as it gives a give balance between including recent and local emissions, but not emissions that have been transport around the globe and become well mixed. The results from our sensitivity tests on the decay rate into the background shows that 9 months provides this balance and choosing a 12 month decay rate does not have much impact on the results.</p> <p>See also our response in Reviewer 1’s General Comment 4 where we provide explanation in the text for the background tracer and the choice of a 9 month decay rate. See Section 2.2, lines 108-115.</p>
19	Section 4: It seems like there is little information overall to verify the seasonality of emissions in various regions, which is critical in determining the SCA. Expand on the discussion of this uncertainty. Where did the assumed initial seasonal emission cycles in the model come from, and what were they based on?	<p>We have added a paragraph in Section 4 to discuss how it is difficult to know which emission sector is driving the change in the seasonal cycle, line 509:</p> <p>“There is some uncertainty in the seasonality of CH₄ emissions and how they change over time in Canada, the Middle East, Europe and China & Japan. The emissions used in TOMCAT were discussed in Section 2.3. Our inversion uses prior information from various emission inventories. The prior emissions that predominantly drive the seasonal cycle are wetland emissions from WetCHARTS model and biomass burning emissions from GFEDv4.1s. These emission estimates have been evaluated in previous CH₄ studies (e.g. Parker et al., 2020 and Liu et al., 2020). These prior emissions are optimised, including their seasonality, when the surface observations are assimilated in our inversion. This means that our emissions used in TOMCAT are optimised seasonally, however it is difficult to disaggregate the emission sectors driving the total emissions’ seasonal cycle in each region.”</p>
20	Line 420: Do these numbers refer to the p-values of the trends?	<p>These numbers are the trend values. We have added units to be clear, line 480:</p> <p>“Europe is the third largest contributor to the decrease in the SCA in the NHL (-1.48 ppb). The TOM_regional JJA and DJF trends in the tracer</p>

		contributions to the NHL from this region are 0.06 ppb year ⁻¹ and 0.02 ppb year ⁻¹ , respectively.”
21	Figures: Figure captions should be expanded to include years of data shown.	Figure captions have been edited to include the relevant time periods.