

Author response to the referee comments and short comments to the paper by Barten et al.: Role of oceanic ozone deposition in explaining short-term variability of Arctic surface ozone

We would like to thank the three anonymous reviewers for their extensive reviews and Owen Cooper and Ashok Luhar for their short comments. All comments are addressed individually starting with the three anonymous reviews and ending with the two short comments. Referee comments are given in *italic*, author response are given in normal font. This document is finalized by a markdown version of the manuscript including all the changes made to the text.

Review #1:

General Comments

In general, the paper is well presented, well written with a sound and detailed introduction, and with appropriate figures and tables. However, at first sight, results seem to be on a low side for Vd-O₃ compared to other results found in the past literature (over the past 20 years or so). Moreover, with relatively little spatiotemporal variation in the High Arctic, for dry deposition velocity (0.012 ±0.002 cm/s), the authors nevertheless claim a greater sensitivity of Vd with respect to environmental factors with COAREG vs DEFAULT. DEFAULT uses a constant for r_s and no variability of surface resistance is allowed. The variability depends only on the aerodynamic and R_b resistances for the latter. Therefore, it is not clear to what the word "sensitivity" and "high variability" refers to in this context for COAREG. For example, the standard deviation of COAREG (0.002) is smaller than DEFAULT (0.003) while the authors claim a greater sensitivity with COAREG. In comparison, other authors (see specific comments and references below) have shown a real and much larger sensitivity and variability than here over the same domain with respect to environmental conditions, for ozone and other gases. For CO₂, many authors have shown a dependence on the square or cubic with windspeed for gas transfer to the ocean while here, the dependency of deposition velocity on windspeed seems small with respect to water-side turbulence and its impact on Vd-O₃. Sensitivity tests with respect to environmental conditions (iodide conc., windspeed, SST, salinity, etc.) should be clearly presented with identification of which environmental factors contribute the most to the variability in COAREG. The authors should also clearly explain the little sensitivity of windspeed for ozone (as compared to other gases such as CO₂, for example). Finally, the originality of the paper is questionable since many other authors have done the exercise of including mechanistic model such as COAREG in ACTM models. Therefore, one may question the science advancement brought by that paper since from the work of recent authors, it becomes obvious that a constant for surface resistance (r_s =2000 s/m) is too high for northern regions (in summer) and this paper is just another confirmation. Finally, the authors question the value of r_s in DEFAULT (2000 s/m) which results in Vd ~ 0.05 cm/s. However, Ganzeveld et al. (2009) stated the following "Solely based on these comparable global annual mean VdO₃ one could draw the conclusion that the commonly applied ConstRs approach (using an R_s of 2000 s m⁻¹) seems to provide a good first-order estimate of global and long-term average oceanic ozone dry deposition for use in atmospheric chemistry and transport models". The presented paper here, seems to contradict this. Please explain and resolve this apparent major contradiction.

We greatly appreciate the detailed review by reviewer #1. Here we would like to respond to some of the more general comments above while we use the specific comments below to address the changes made in the manuscript. First of all, this paper appeared to show ambiguity regarding the use of "very sensitive" or "high variability". In this context, we mostly refer to the "high sensitivity" of the model to the representation of oceanic O₃ deposition in simulating surface O₃ concentrations (whether the standard Wesely approach or the process-based approach is applied). We hope that by showing the sensitivity of the COAREG schemes to environmental factors (Fig. B1) the interpretation of Fig. 3 becomes more clear. This is addressed in more detail below with the response to the other comments.

Here we would also like to respond to the apparent contradiction with Ganzeveld et al. (2009). Indeed, based on only comparing the global mean V_{d,O_3} with the COAREG model in EMAC as well as checking the overall changes in the global annual deposition and O_3 burden, it seemed that the constant r_s approach was providing comparable results as the COAREG implementation in EMAC in 2009. Now, this further detailed and focused study on Arctic O_3 and the recent work by others on application of these mechanistic representations (e.g. Luhar et al. (2018), Pound et al. (2019)) indicate that the constant r_s approach is not applicable for analyzing Arctic O_3 data on shorter timescales.

Specific comments

1) High variability/sensitivity of V_d-O_3 over Arctic waters

-In the introduction, the authors correctly mentioned the sources of variability of dry deposition over oceans (lines 63-73). From this, the reader would expect a much larger variability than that of DEFAULT. However their results shown in the paper (Fig. 3c and table 1) rather indicate a rather small variability around the mean $V_d-O_3 = 0.012$ cm/s. In fact, according to Table 1, the absolute variability in COAREG is actually less (0.002 cm/s) than that in DEFAULT (0.003 cm/s). In the conclusion, the authors repeat (line 373-375); "we show that Arctic surface O_3 concentrations are very sensitive to the representation of O_3 deposition". This claim is not supported from the results presented. Inter-seasonal variation of dry deposition velocity was shown to be greater than the spatiotemporal variation over the domain shown in the presented paper (compare with Figs 3a,b and Fig. 9 of Ganzeveld et al. 2009). Similarly, in other studies, the variability of V_d-O_3 over oceans seem much larger (such as In-Bo Oh et al. 2008, Chang et al., 2004). Chang et al. (2004) (their Fig. 2) reports a large variability in ozone deposition velocity observations over the world oceans and a large sensitivity to windspeed (V_d about in the range 0.015 - 0.07 cm/s; mean about 0.03 ± 0.015 cm/s from their Fig. 2), Again, how do you reconcile that with your results: $V_d = 0.012 \pm 0.002$ cm/s ?

The variability (indicated by the standard deviation of 0.002 cm s^{-1}) given in Table 1 represents combined spatial and temporal variability (combination of Fig. 3 a,b and Fig. 3c) of the simulated O_3 deposition velocities and therefore does not represent the variability with respect to wind speed or Iodide concentrations separately. In absolute terms, this variability (0.002 cm s^{-1}) is indeed lower than the variability of DEFAULT (0.003 cm s^{-1}). However, since the magnitude of the mean deposition velocity is an order of 4 smaller compared to DEFAULT it is larger in relative terms.

Statements made in the manuscript that mention 'high variability' or 'very sensitive' deal with the sensitivity of the surface O_3 concentrations to the representation of the ocean-atmosphere exchange and not to the variability/sensitivity of the deposition parameterization itself. In other words, which deposition routine (Wesely or COAREG) is used, affects the simulated surface O_3 concentrations and the comparison with observations which adheres to the main goal of the paper. The goal of this paper is not to develop or optimize the ocean-atmosphere exchange routine, but rather apply such a routine to improve simulations of the short-term spatiotemporal distribution in surface O_3 . To avoid confusion, we have removed all instances of mentions of 'high variability' or 'very sensitive' throughout the paper.

The deposition velocities presented in Table 1 ($0.012 \pm 0.002 \text{ cm s}^{-1}$) show a slightly lower magnitude and similar variability to In-Bo Oh et al. (2008) Table 2 Case 3 ($0.0160 \pm 0.0015 \text{ cm s}^{-1}$) which is their case that includes the removal of O_3 by Iodide. In that study, typical iodide concentrations are in the order of 100-200 nM (up to 400 for coastal waters) whereas in our study we have typical iodide concentrations of 30-130 nM (Fig. C1) which can explain the lower magnitude of ozone dry deposition velocities.

We agree that there is need to show the sensitivity to environmental factors to clarify the (lack of) variability in the simulations. More information can be found in the reply to the next comment.

2) Sensitivity of environmental factors

*- The sensitivity with respect to wind is unclear in the paper. Wanninkhof (1992), McGillis et al. (2001a,b) have shown a strong dependency (U^{**2} or cubic root U^{**3} with windspeed) for air-sea gas*

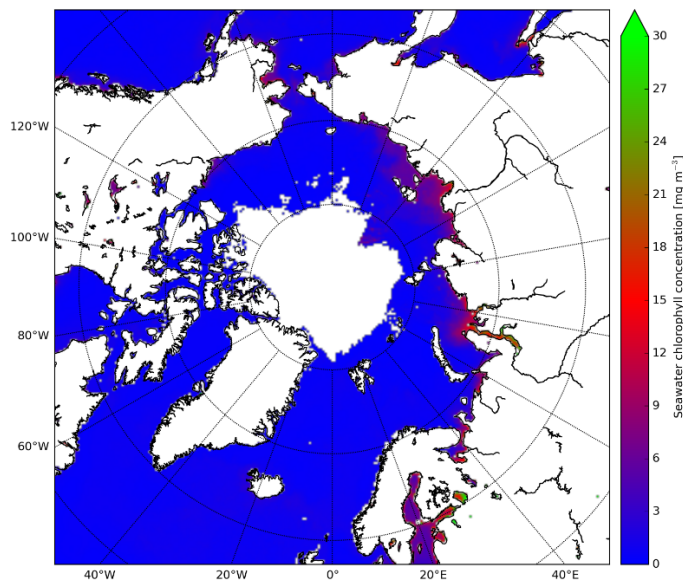
exchange. Please comment more clearly about the sensitivity vs windspeed. Moreover, in the study presented, it seems that the biogeochemistry spatiotemporal changes do not impact much $V_d\text{-O}_3$. For example, other authors have clearly really demonstrated a large variability (e.g. Table 2, Fig 3a,b, Fig. 4 and Fig.8, of Genzeveld et al. 2009). Helmig et al. (2012) provide a large variability for $V_d\text{-O}_3$ from 0.01 to 0.1 cm/s (as mentioned by the authors Barten et al. 2020 in line 58 of their paper). Therefore, the variability shown by the authors here again appears much smaller for Arctic regions ($V_d=0.012 \pm 0.002$ cm/s) than the above authors despite the authors claim high sensitivity. Please explain. My understanding is that the intra-annual amplitude of dry deposition for ozone is large at high latitudes (e.g. Fig.3,b and Fig.4 of Genzeveld et al. 2009). The authors should state clearly state that the variability for $V_d\text{-O}_3$ presented applies only in summer and under special conditions so that readers would not be tempted to extrapolate the results to other seasons or to lower latitudes, or anywhere else. In the literature, the inter-annual variability is up to 0.15 cm/s in the North Atlantic. The authors reports a summer variability of only 0.002 cm/s for dry deposition velocity. - According to Clifford et al. (2008), ozone deposition velocity is up to 0.1 cm/s in high chlorophyll (found in coastal waters in North Atlantic and Arctic in concentration up to 3-4 mg/m³). This dry dep. velocity range seems to agree with Chang et al. 2004 (range 0.015 cm/s to 0.07 cm/s), although the latter study dealt with lower latitudes. What are the levels of chlorophyll in your domain here ? The authors have to explain more clearly why they divert drastically from past literature and why chlorophyll-alpha is not important here. Moreover, Gallagher et al. (2001) proposed an average value of surface resistance of 950 s/m (corresponding to about $V_d = 0.1$ cm/s) for coastal UK. Do you obtain similar values ? if not, this means significant sensitivity to u^* greater than shown in the paper here. Chang et al (2004) has shown a factor of 5 for deposition velocity of ozone with windspeed increasing from 0 to 20 m/s. Similarly, In-Bo Oh et al. (2008) reported values of surface resistance decreasing rapidly for $[I^-]=100\text{nM}$ from 5000 sm^{-1} at zero wind speed to about 1000 sm^{-1} at 20 m/s windspeed (their Fig. 4). For turbulent air (aerodynamic resistance negligible), this corresponds to dry deposition of 0.02 to 0.1 cm/s respectively. Therefore, I have some trouble reconciling this with the conclusions of the paper presented here. In any cases, authors should not claim high sensitivity for summer in Arctic region but rather a large discrepancy with the DEFAULT constant value for r_s vs COAREG with small variability around the value 0.012 cm/s. More importantly, the authors should present a table showing the sensitivity of each environmental conditions in COAREG and show the results of sensitivity tests to support and clarify their claim. I suggest that the authors first present a table describing basic simulated statistics about environmental conditions, windspeed, SST, iodide conc, salinity, aerodynamic resistance (R_a), boundary layer resistance (R_b), surface resistance, etc. to better understand the link with V_d in the Arctic and O_3 mixing ratio and also provide sensitivity tests (as already discussed above).

We agree that providing such a sensitivity analysis benefits interpretation of the shown results. Therefore, we have performed additional simulations to test the sensitivity to the environmental factors wind speed, SST and oceanic iodide concentrations (Figure B1) and have also shown the spatial distribution of I_{aq} used in the simulations (Figure C1) to show typical summer Arctic I_{aq} concentrations. For typical Arctic summer $I_{\text{aq}} = 60$ nM the sensitivity to wind speed is rather low (0.008 cm s^{-1} at 1 m s^{-1} winds to 0.015 cm s^{-1} at 15 m s^{-1} winds). At very low wind speeds (< 3 m s^{-1}) the increase in aerodynamic resistance poses another restriction on O_3 exchange which is not included in e.g. In-Bo Oh et al. (2008) Fig. 4 that shows the relation between wind speed and surface resistance. The role of solubility (Fig. B1,b) seems to be compensated by the role of chemical reactivity (Fig. B1,c) for typical Arctic SST's and I_{aq} .

As an addition to Fig. B1, that shows the sensitivity to environmental factors, we have included in Section 3.1 a comparison with other literature (e.g. Chang et al. 2004, Oh et al. 2008, Luhar et al. 2017) to put the results of our simulation in perspective of other observed and simulated temporal variability in V_{d,O_3} . The variability of O_3 deposition is often represented on the global scale and/or including the variability over the different seasons. In this study we show the variability of O_3 deposition in one month with nearly constant I_{aq} concentrations making the variability mostly determined by changes in wind speed. As mentioned before, the simulated O_3 deposition velocity and variability presented in Table 1 (0.012 \pm 0.002 cm s^{-1}) seems to show similar variability to In-Bo Oh et al. (2008) Table 2 Case 3

($0.0160 \pm 0.0015 \text{ cm s}^{-1}$) which also performed a simulation over 1 month (21 July-20 August 2005) over the Gulf of Mexico. Furthermore, Luhar et al. (2017), e.g. Figure 7, showed a large (observed and simulated) sensitivity to wind speed for some measurement campaigns (e.g. TexAQS06 and GOMECC07) in contrast to a low sensitivity for e.g. GasEx08 in the relatively cold Southern Ocean. Furthermore, as indicated by Luhar et al. (2017) the O_3 deposition velocities in one-layer schemes is overestimated by a factor of 2-3 due to the enhancement of the interaction between chemical reactivity and waterside turbulent transport (Luhar et al. (2017) Fig. 6). The relatively newer two-layer schemes seem to more accurately represent O_3 deposition flux measurements and the dependency to SST and wind speed (Luhar et al. (2017) Fig. 7).

For the case of chlorophyll- α we have included in the Discussion section a sensitivity analysis to the Chl-O_3 reaction using MODIS chlorophyll- α for the oceanic boundary condition. For these Arctic summer simulations the role of Chlorophyll seems to be limited in comparison to the role of I_{aq} . For open oceans typical chlorophyll- α concentrations are $< 3 \text{ mg m}^{-3}$ (see Figure below, not included in the manuscript). As indicated in the discussion, we only found a slight increase in O_3 deposition to waters having chlorophyll- α concentrations $> 25 \text{ mg m}^{-3}$. We have added extra information on typical chlorophyll- α for this Arctic summer case.



3) Originality/added value

-Overall, I did not find that the results are of significant impact and have substantial originality vs existing literature. Other authors have modified ACTM models with mechanistic dry deposition scheme over water (Pound et al., 2020; Helmig et al. 2012; Fairall et al., 2011; Luhar et al., 2011; Coleman et al., 2010; Ganzeveld et al., 2009; In-Bo Oh et al., 2008; etc.). Perhaps, the authors should clearly provide a statement discussing the added value to the existing literature. The case presented seems a special case where there is a limited role of water-side turbulence, iodide variation, impact of halogen chemistry influence, chlorophyll and organic matter, etc. A comparison of winter versus summer case would have been more interesting.

We are aware that this is not the first study that has coupled a mechanistic dry deposition scheme to water bodies to an ACTM. The goal of this study is not to improve these dry deposition schemes nor to quantify the impact of all driving factors (e.g. waterside turbulence, solubility, reactivity with Iodide but also e.g. DOM, etc.) in different seasons. Rather we illustrate that such a mechanistic representation in ACTMs is needed not only to an improved representation of the magnitude of the O_3 deposition sink term (Fig. 3) and long-term (e.g. monthly-mean) surface O_3 concentrations in the High Arctic and above oceans (Fig. 4), but also to better represent the observed short-term temporal variability in surface O_3 (Fig. 5 and Fig. 6). To the authors' knowledge, this is the first time that such an evaluation of the short-

term surface O₃ variability with respect to oceanic O₃ deposition is performed and also compared with a large dataset (25 stations) of Pan-Arctic hourly surface O₃ observations.

Line 35: Ozone has also significant impact on destroying materials by oxidation, see <https://www.sciencedirect.com/science/article/pii/S1352231095004076> or https://www.worldscientific.com/doi/abs/10.1142/9781848161283_0009 I think a word about impact on materials should also be mentioned there for completeness.

We have added the reference to Lee et al. (1996)

Line 43-44: Changes in deposition velocities (linked with changing meteorological and oceanic conditions, stomata closure, droughts, etc.) may also contribute to these trends in mid-latitude.

We have added mentioning of changes in O₃ deposition to vegetation in the introduction.

Line 57-58. Over oceans, Clifford et al. (2008) suggest values of Vd-O₃ up to 0.1 cm/s, Chang et al. (2004) had Vd-O₃ in the range: 0.015 cm/s to 0.07 cm/s. Gallagher et al. (2001) had Vd-O₃ up to 0.1 cm/s near coastal waters. Perhaps these references deserved to be mentioned for oceans and coastal waters as well to give more background about the real variability of Vd-O₃.

This statement in the introduction is to give the reader an idea of the order of magnitude and range (typically between 0.01 and 0.1 cm s⁻¹) of O₃ deposition to oceans and to mention that it is a relatively slow process compared to O₃ deposition to vegetation. We have added the references to the different papers.

Line 62. It would be very interesting for the reader to know where this value of r_s cm/s (DEFAULT) comes from. Ganzeveld et al. 2009 seems to agree with the constant for northern latitude ($r_s=2000$ s/m) as stated above.

This constant surface uptake resistance for water bodies originates from Wesely (1989) and is therefore still commonly applied in most ACTMs. We have included the explicit mention of the origin in the text.

Line 93-96. The reaction ozone + iodide is a fast reaction why it doesn't affect short time scales as well ?

This statement is based on the variability of the drivers of O₃ deposition. Regarding wind speed, large variability can occur within ~1 or 2 days by e.g. passing of a dynamic system (low pressure area). On the other hand, temporal variability of SST and I_{aq} is rather slow and is occurring more at timescales of weeks/months. The O₃-I_{aq} interaction is indeed occurring at fast timescales, however, the temporal variability makes changes/trends in I_{aq} affect the longer-term variability (e.g. seasons) in oceanic O₃ deposition.

Line 125. The choice of the period is well supported according to the authors (end of summer 2008). However, the reader should be reminded that the conclusions of this study only strictly applies for summer 2008. Waves height are highly variable in the north Atlantic and therefore the water-side turbulence in other seasons. Under high chlorophyll conditions (as seen by MODIS instrument), algae bloom, etc., the fate of ozone is possibly more in other seasons. Therefore, there will be cases when the net dry deposition would be much higher than 0.012 cm/s. The authors should not leave the reader under the impression that $vd=0.05$ cm/s currently used in model is too high everywhere in any seasons. I wonder about any contribution of ozone subsidence for higher altitudes in the High Arctic ?

Also based on another comment we have added in the discussion limitations of this study including limited potential to extrapolate these results to other seasons/latitudes. By also including the I_{aq} distribution in Figure C1 the results of this study can be put in perspective in terms of chemical reactivity of the Arctic ocean in summer.

One major motivation to focus this study on evaluation on August 2008 was limited data availability. Measurements of O₃ at many of the stations are indeed available whole year round but High Arctic O₃ concentration measurements were mainly limited to the ASCOS campaign, August 2008. But we have extended the discussion to further stress that the apparent very small VdO₃ for this one month

evaluation period cannot be deemed being representative and that further evaluation with the MOSAiC 1-year campaign observations, including O₃ fluxes and oceanic Iodide will further provide an insight in annual variability in high Arctic V_{d,O3} and its impact on O₃.

The model accounts for O₃ subsidence by solving besides horizontal transport also the vertical transport of chemical species. We have not found a clear indication of strong subsidence O₃ affecting surface concentrations in this period.

Line 125-126. Is halogen chemistry limited only to spring time ?

Halogen chemistry is not solely limited to springtime. However, in August/September the contribution of halogen species on Arctic surface O₃ is much more limited compared to the period February-June (see Yang et al. 2020, Figure 3). We have added the reference to Yang et al. (2020) in the manuscript accordingly.

Line 154. "Extension for a two-layer scheme vs Fairall et al. 2011". The authors should provide briefly more details on how these two layers are structured for the benefit of the reader.

Also based on comments by Reviewer #2 and Ashok Luhar we have included the formulation of the air- and waterside resistance terms in Appendix B.

Line 158. It is not clear why chlorophyll-alpha from MODIS as proxy for iodide and organic matter is ignored. Such proxy has been used with success in previous literature (In-Bo Oh et al., 2008). A good linear correlation was found between iodide and chlorophyll-alpha. The advantage of using MODIS is to obtain a very good spatial coverage (not the case with ground point measurement).

We have included the reference to Oh et al., (2008) and the mentioning of Chlorophyll-a derived iodide concentrations for completeness. In the Sherwen et al. (2019) product, Chlorophyll-a (but also e.g. nitrate and SST) has been used as a predictor to derive the oceanic Iodide concentrations. Satellite derived chlorophyll-a has therefore been (indirectly) included in this study in the sense that it is integrated in the Sherwen et al. (2019) product. In the discussion we indicate the further use of Chlorophyll-a also as a proxy for DOM being an additional potentially important reactant including a sensitivity analysis (see also the reply on 2) Sensitivity of environmental factors)

Line 173. About machine learning (ML) approach. It needs more details. ML is a generic term. Which ML was used ?

We have added ' , namely the Random Forest Regressor algorithm (Pedregosa et al. (2011))' to the manuscript. We have included this in the manuscript. This method used the top-10 performing regression models in an ensemble prediction.

Section 2.2.1 and 2.3 Ozone could be destroyed by chemical reaction with snow. Not clear how it is taken into account in the study presented. Please provide more details here or refer to a discussion later in the paper. The authors do not provide clear scientific reasons to why they decrease V_d-O₃ for snow/ice from 0.03 to 0.01 cm/s (although it fits better the observations). Writing "Based on Helmig et al." is not sufficient . Please add-up a bit more details.

We have now included the recent review by Clifton et al. (2020b) summarizing observed O₃ deposition velocities to snow similar to Helmig et al. (2007a) but also including more recent measurements. Clifton et al. (2020b) also summarized that accurate (process-based) modelling of O₃ deposition to snow requires better understanding of the underlying processes. We think that introduction of process-based O₃ deposition to snow in WRF would currently introduce many more uncertainties also related to limited spatiotemporal observations of some of the dependencies (e.g. bromine, formic acid, ...). Therefore, we have decided to apply the 'best estimate' of surface uptake resistance to snow by Helmig et al. (2007a). We have indicated in the discussion that process-based modelling of O₃ deposition is currently hampered by multiple factors: "Furthermore, we have reduced the deposition to snow and ice".

Line 238-239. Variability of O₃ deposition of 20% in turbulent transport looks small. Other authors have found a factor of 5 with windspeed (Chang et al. 2004).

Also based on the comment "2) Sensitivity of environmental factors" we have included in the results Section 3.1 a comparison to magnitude and variability of similar mechanistic representations applied in other studies.

Line 160. Nitrate is used as a proxy for iodide concentration. Chlorophyll-alpha is another proxy available from satellite (MODIS). Again, why not considering satellite measurement of chlorophyll since the spatial coverage is much better? Anyways, a comparison of the two methods would be of interest.

See reply to comment "Line 158. It is not clear why chlorophyll-alpha from MODIS...".

Line 226. VD increases over warmer water (Fig. 4) but the solubility of ozone and other gases (such as CO₂) generally decrease with increasing sea surface temperature. Therefore, in principle, this produces less ozone uptake by ocean if everything else is equal. Your results show the opposite: increase from 0.01 to 0.018 cm/s from cold to warm waters. What is the impact in % of the solubility effect on Vd-O₃ vs other factors. Perhaps the effect of iodide counteracts effect of solubility. Please discuss.

This indeed shows one of the compensating effects in the oceanic O₃ deposition process. We had indeed misworded the role of SST and Iodide in this Section. We have updated the text and included in Fig. B1 the sensitivity to SST (Fig. B1b) and Iodide (Fig. B1c).

Figure 3b,c. -The result of the authors show rather low deposition velocity (0.012 cm/s) with relatively low variability (0.002, i.e. less than 20% variability). In fact the variability (e.g. Fig 3c) is less than the default (the latter having a surface resistance taken as constant). Compared to the literature, the results obtained by the authors are among the lowest Vd and among the lowest variability found. Please indicate which authors, and which paper would support the results found? For example, Coleman et al. (2010) using different scenarios computed much higher VD = 0.0547 cm/s (for iodide conc. of 100 nM) for the North Sea. Ganzeveld et al (2009) shows a worldwide map of deposition velocity of ozone over oceans for January and July. The simulation for summer (their Fig. 3b) shows a minimum of 0.025 cm/s (range 0.025-0.045 cm/s) for the domain of the study presented here for dry dep ozone. Moreover, although the location is significantly different, Chang et al. (2004) mentioned a high variability of VD (ozone) of at least 50% (compared to less than 20% in the authors study). Therefore, a question arises: what particular conditions of Arctic at that period of the year 2008 in summer would produce such low variability and low deposition velocity?. I understand iodide conc. is low, in the context of the paper presented, moreover the authors neglected halogen chemistry, etc. but still, I think the authors should explain better why their Vd are so low and their variability not so high as well although the authors claim a high sensitivity. I also suggest that Fig. 3c should show the time series at various locations not only at a single one.

Please refer back to the response on one of the main comments namely: "2) Sensitivity of environmental factors". We have changed Fig. 3 to show the NUDGED and COAREG simulations instead of the DEFAULT and COAREG simulations. Adding various locations to Fig. 3c made the figure very messy and we have decided not to include this in the manuscript also because now the various sensitivities also to wind speed are shown in Appendix B.

Figure 4. Concerning differences between CAMS and COAREG over land: could it be explained by modification of the Wesely scheme (1989) over land to take into account LAI (i.e. bug in Wesely, 1989; see correction in Val-Martin et al, 2010)? I suspect one model has integrated the Val-Martin's correction and the other not (e.g. to explain differences over Scandinavia, Russia and Northern Europe between the two models CAMS and COAREG). Please comment or check on this.

Also based on comments by Reviewer #2 regarding the role of CAMS in this manuscript we have removed CAMS from Figure 4. However, we would like to shortly comment on the deposition schemes in the WRF and CAMS models. The CAMS model uses the SUMO (Michou et al., 2004) dry deposition calculation whereas WRF uses the Wesely scheme which is often updated with recent advances/bug

corrections such as those by Val-Martin et al., (2010). We have included a statement on the different representation of deposition in Sect. 3.3.

Line 273. Vd (ocean) is about 0.012 cm/s and over snow/ice about the same. i.e. 0.010 cm/s (small gradient) Therefore, why is there a sharp gradient from Greenland vs sea (Figure 4). Authors should perhaps say a word about it (altitude effect, accumulation of ozone over Greenland, descent of ozone from higher altitudes over Greenland, etc. or any other reasons ?).

We have included a statement on the higher simulated O₃ above Greenland due to the altitude effect.

Line 275 and Figure 4d. The COAREG distribution is closer to a Gaussian distribution than that of DEFAULT. I think it is worth to briefly mention it.

Based on comments by other reviewers we have removed this panel from the figure.

Table 2. Note that bias, MAE and R are somehow redundant metrics (show similar information). I think the authors should consider adding up another metric which is entirely orthogonal to bias such as the standard deviation of O-P (Observations minus model prediction) or any other metrics showing the random error. Bias and MAE both show systematic errors (i.e. Table 2 either give information on the systematic error or on the degree of correlation). See Chang and Hanna (2004) for metric redundancy.

We have included in Table 2 the standard deviation of O-P and have removed the Bias. The presented results are similar including this new metric and removing the Bias. The COAREG simulation outperforms the NUDGED simulation at the 6 High Arctic sites both in terms of systematic error, random error and degree of correlation. We think that R is supplemental to Bias/MAE as there can be a perfect degree of correlation (R=1) but still the data can have a large bias (or MAE in that sense). Therefore we have left R in the paper to indicate the degree of correlation and the ability of the model(s) to capture the short-term variability.

Line 374. What is your criteria to conclude about the high sensitivity ? To which environmental conditions Vd-O₃ is very sensitive: windspeed, temperature, salinity , iodide concentration ? . Again, I would suggest providing evidence of sensitivity by making sensitivity tests and showing the results as a form of a Table. DEFAULT was driven by a constant which is too high and likely not applicable for arctic regions in summer. COAREG does not use this constant but shows little variability around the mean, i.e 0.012 ±0.002. Please re-word or add specific evidence for high sensitivity.

This statement is based on the actual representation of oceanic O₃ deposition on simulated surface O₃ concentrations (e.g. Fig. 5 and Fig. 6). To avoid ambiguity we have removed 'very' sensitive but also added explicitly that this sensitivity is highest for the High Arctic and coastal sites. As mentioned before, to clarify the sensitivity of COAREG to environmental factors we have included Fig. B1 in the manuscript.

Line 496. "It corroborates findings of which study on global scale" ? The authors should give references to that statement. As mentioned above, values shown for Vd are lower w.r.t to previous literature in general. Conclusion: I think somewhere, the author should comment about the need for open ocean measurements (for iodide, DOM, halogen, ozone, weather variables and other relevant environmental variables) and/or of flux measurements. These measurements are needed to validate models and quantify better open ocean chemistry near-surface. Observations shown are limited and conclusions should be taken with care. Authors should recognize the limitations of their study (no halogen chemistry included; results cannot extrapolated to other seasons, lower latitude, etc.).

We have added references to this statement. We agree that there is need for open ocean measurements to reduce the uncertainty both in terms of driving factors (e.g. Iodide, DOM, etc.) as well as direct flux measurements to better validate and constrain these regional and global modelling setups. We have added a section in the Discussion to address the need for additional measurements.

Technical corrections

Line 12 and 465: "we have coupled the Coupled-": redundant words.

Changed 'coupled' to 'integrated'. "Coupled" here refers to the full name of the COAREG algorithm.

Line 29: "is used" -> "be used"

Changed to 'should be used'.

Line 36: ozone lifetime differs according to NOx source proximity or altitude. Should indicate that it is the corresponding lifetime in the free troposphere (not near surface or in the upper troposphere or stratosphere which differs substantially).

Added 'in the free troposphere'.

Line 194 and 488. (sea-)ice -> sea-ice

Changed to 'sea-ice'.

Line 225 and 227 deposition -> deposition velocity (figure 3 deals with deposition ve-locity, not deposition)

Added 'velocities' in line 227

Line 234. Up to 8% reduction ? Seems a bit small to me. Say $R_a = 2000$ (under temp. inversion), $R_s = 2000$ (default) , -> $V_d = 0.025$ cm/s a 50% reduction. Please verify.

This statement is based on the simulated output of the DEFAULT run. Only at very rare occasions the $V_{d,03}$ drops below 0.04 cm s⁻¹ ($r_a = 500$ m s⁻¹) also visible in Fig. 3. The simulated wind speeds above oceans hardly drop below 3-4 m s⁻¹ preventing strong temperature inversions above oceans. Whether this is realistic or not is hard to say also because the AMSR-E satellite retrievals have a large error at these low wind-speeds. We have edited the text to indicate the lower limit of simulated $V_{d,03}$ in the DEFAULT run (0.4 cm s⁻¹).

Line 239-240. Reduction from 0.03 to 0.01 cm/s gives a reduction 66% , not 30% !

Changed to '66%'.

Line 258. I suggest re-wording "We find a limited effect..." -> "As expected, we find a limited effect.."

Added 'As expected,' .

Line 278-279. Improve in what sense ? model predictions scores improvement ? Line 278. Improve short-term -> increase the short-term

We have added explicit mention of 'model prediction scores'

Line 280 such a oceanic -> such an oceanic

Changed a to 'an'.

Line 364,372, 495. role -> impact

We have changed role to 'impact' at several occasions in the text

Line 374. address or include ? not both.

We have removed 'include'

Line 403-420. Much of the stuff should go in the Methods section 2.

We have moved a significant portion (when discussing the different I_{aq} parameterizations) to the Methods section.

Line 439. meteorolog -> meteorology

Changed to 'meteorology'.

Line 478-480. This is not clear. What is dominant, sensitivity to iodide, solubility, temperature or windspeed ? Showing a table with sensitivity tests would be appreciated.

As mentioned in previous replies we have included the sensitivity to Wind speed, Solubility (SST) and Iodide.

Line 483, 484. I suggest you replace % -> reduced by a factor of 3.4 (ocean) and 2.6 (ice).

We have added the factors instead of the percentages

Line 496. It corroborates which findings ? (needs a reference)

We have added references to this statement.

Author contributions: what is the precise role of Maarten Knol in the study ? Please specify.

All authors contributed to writing the manuscript. We have explicitly added this in the Author Contributions

Review #2:

The authors revise the ozone dry deposition scheme in WRF Chem (now, 'COAREG'). They perform several WRF Chem simulations of August 2008. First, the authors perform a default simulation. Finding that there needs to be nudging to observed winds, they perform a nudging simulation and a deposition+nudging simulation. The paper would be much stronger (and adhere to its goal of investigating the impact of ozone deposition) if the authors focused on the comparison between the nudging simulation and the deposition+nudging simulation, instead of comparing the default and the deposition+nudging simulations. The authors hypothesize that the original ozone deposition scheme in WRF Chem (Wesely) overestimates the magnitude of and underestimates variability in ozone deposition velocity (V_d) over the ocean. They also hypothesize that the magnitude of ozone deposition velocity over snow and ice is overestimated. In general, I think V_d over Arctic land, ice, and ocean are all very uncertain in terms of magnitude and variability. I would like to see this mentioned in the abstract and conclusion. For example, can the authors really say that it's V_d variability over the ocean that driving the improvement in ozone variability when V_d variability over snow and ice is uncertain and likely not represented accurately? The bulk of the paper is about the impact of COAREG on the mean bias of ozone, both in terms of spatial and hourly scales. The authors could do a better job at indicating whether COAREG improves spatial and hourly variability (i.e., be more quantitative). The title, which should be slightly revised (see below), reflects the strength of the paper, which is really in Figure 6 where the authors illustrate that COAREG improves short-term variability in surface ozone at 5/6 sites in the high Arctic and during ASCOS. However, one thing that is unclear is how the authors chose the six sites (out of 25) to highlight in Figure 6. Are these just the sites that COAREG shows a clear improvement at? The paper would strongly benefit from further analysis of short term variability in ozone deposition velocity in COAREG: what's driving the variability in deposition velocity, in particular in periods of better agreement or disagreement with surface ozone? Is it that day-night differences are better captured? Day-to-day variability? Synoptic scale variability? Currently the discussion of Figure 6 seems a bit anecdotal/random. Overall, I recommend major revisions. I think for this paper to have sufficient novelty for publication in ACP the authors need to expand on their analysis of short-term variability at high Arctic sites. The paper is generally well written and clear but can be very wordy and long-winded.

We thank reviewer #2 for her/his extensive review and agree that tackling the raised remarks will help to substantially improve the manuscript. Here, we give a general response to the addressed points and more detailed responses can be found below. We have adjusted the setup of the paper to focus on the comparison between the NUDGED and COAREG runs instead of the DEFAULT and COAREG runs. We hope that the revised structure of the paper by changing Fig. 3, Table 1, Fig. 4, Table 2 and Fig. 6 better reflects the main objective of the manuscript. We have also revised Section 3.4 (Figure 6) to put more emphasis on the short-term temporal variability in observed and simulated surface O_3 . At multiple instances we have reduced the length of sentences and removed paragraphs from the discussion that were out of context to reduce the already lengthy discussion.

Title: perhaps should be revised to 'Role of oceanic ozone deposition in explaining short-term variability of surface ozone at high-Arctic sites'

We agree that the title should be revised to emphasize the major point we want to address with this manuscript and have revised the title following your suggestion.

The authors say throughout that this is a 'preparatory' study for MOSAiC, but I don't think this does much for the paper. It's not compelling and feels inappropriate to include in a paper. For every field campaign there is a lot that goes into preparations and forecasting, but this doesn't mean it merits publication.

We have removed the explicit mentions of this manuscript being a preparatory study at several occasions in the text (Abstract, Introduction and Conclusions) also because this manuscript does not solely serve as a preparatory study but mostly to address issues with representing short-term spatiotemporal variability of surface O_3 related to ocean and sea ice deposition. However, in the discussion we bring up

the notation that further evaluation of the role of ocean (and snow-ice) deposition beyond that presented for the month of August will be the next step.

Also, I'm not sure the utility of including CAMS or what I should be taking away from this analysis. Perhaps including CAMS and the default simulation is really for documentation for MOSAiC, but unless the authors can frame the analyses in a more compelling way, then they shouldn't be included here.

The main reason we are also including CAMS in the comparison is that it is a product that is widely used for air quality assessments, long term changes and trend analysis in e.g. O₃ or to constrain regional scale models such as WRF-Chem. Therefore, it is important to understand how CAMS performs also in terms of Arctic surface O₃ forecasts to e.g. indicate where CAMS is performing well (or not). We agree that the comparison with CAMS might have been overdone in the manuscript as the main goal is to address the role of oceanic (and sea ice) O₃ deposition on short-term variability of Arctic surface O₃ and have therefore limited the comparison with CAMS to solely the comparison with hourly surface O₃ in Sect. 3.3 and Sect. 3.4.

The authors are missing ozone flux and deposition velocity constraints from Toolik, Alaska (Van Dam et al. 2016 10.1002/2015JD023914). Please compare how WRF Chem performs. This may signal as to whether terrestrial Vd also needs to be adjusted.

Also based on comments of another reviewer we have added a section in the discussion that addresses potential issues with land deposition and have compared the magnitude of the land deposition flux to bare soil with the observed fluxes from van Dam et al. (2016). However, a detailed analysis of these fluxes is out of scope for this manuscript.

Authors need to revise their use of the term 'background': their usage is incorrect throughout the paper. See Jaffe et al. 2018 <https://doi.org/10.1525/elementa.309>

We apologize for being unaware of this strict definition of 'background' O₃. We have changed the mentioning of 'background concentrations' manuscript to '(lower-)tropospheric concentrations', 'surface O₃ concentrations' or removed 'background' accordingly.

The authors need to more clearly what COAREG is/does. Which variables does it ingest from WRF? What parameters or sub-parameterizations are used?

Also based on the short comment by Ashok Luhar we have added Appendix B that describes the formulation of the air- and waterside resistance terms and gives an overview of the sensitivity to environmental factors.

Specific comments

Line 5-6: with respect to 'is also overestimated': the authors haven't yet provided an indication of whether ozone deposition to the Arctic Ocean should be over or underestimated, only that it shouldn't be constant. Given that this overestimate is discussed through the rest of the abstract, please give your hypothesis as to why it is overestimated here.

The statement 'is also overestimated' is based on previous global modelling studies and relies especially on the low reactivity of the Arctic ocean as was already included. We have changed the line to: "We hypothesize that O₃ deposition to the Arctic ocean, having a relatively low reactivity, is overestimated in current models with consequences for tropospheric concentrations, lifetime and long-range transport of O₃."

Line 9: I don't know what MOSAiC is

Based on one of the comments given above we have removed the statements including MOSAiC in the abstract. Furthermore, we have introduced the full name (Multidisciplinary drifting Observatory for the Study of Arctic Climate) at the first instance it is introduced in the text.

Line 16: I don't know what ASCOS is

We have included the full name (Arctic Summer Cloud Ocean Study) in line 16.

Line 30: 'can be' is a bit of a stretch here: these observations haven't even been made.

Based on one of the comments given above we have removed the statements including MOSAiC in the abstract.

Line 39: Observations of background ozone are not possible

We have removed the term 'background' here.

Line 49: Is Hardacre et al. 2015 the correct reference here?

We have updated the references in Line 49.

Line 58: reference for V_d, O_3 up to 2 cm/s?

Included reference to Fan et al. (1990)

Line 59: Hardacre et al. 2015 should be cited here as well

We have added the reference to Hardacre et al. (2015).

Line 78: 'the mechanistic representation in Pound et al. (2019)' instead of just 'this mechanistic representation'

Changed to 'the mechanistic representation in Pound et al. (2019)'.

Line 76-80: I'm not sure how this is a 'for instance' of an important feedback mechanism

We have restructured the paragraphs here to make the flow of the text better

Line 86: do the authors mean 'evaluating with monthly mean . . . observations'?

We have changed 'using' to 'evaluating with'

Line 89-91: not sure what why sub-monthly concentrations will help constrain the "background" concentration. . . please elaborate

Reading again the statements regarding evaluation of sub-monthly O_3 concentrations we also realized that these do not clearly reflect what we wanted to express and have removed those statements

Line 91-92: I think the authors need to make a stronger argument that simulated ozone deposition evaluation relies on evaluation of high frequency temporal variability O_3 observations

Due to the lack of Arctic ocean-atmosphere O_3 deposition flux measurements this evaluation relies on the evaluation of a wide network of surface O_3 measurements. We have adjusted in the text to make more clear that the evaluation is hampered by the lack of flux observations.

Line 96: reference for iodide controlling longer term changes in V_d ?

This statement is based on the different timescales of variability of the drivers of waterside turbulent transport and chemical enhancement. Where the drivers of waterside turbulent transport (mostly wind speed) have a strong day-to-day variability, the variability of Iodide is more on the monthly timescales. We have added an explicit mention of the monthly variability in Iodide to drive the more long-term (weekly-monthly) changes in O_3 dry deposition.

Figure 1: I don't see a drifting path for the ASCOS campaign. Can you make the line more bold? Can the authors use different colors for the sites that show whether they are high arctic vs. terrestrial vs. remote sites?

The drifting path of ASCOS is quite short for the time of the simulation and is therefore not directly visible from a distance. We have made the drifting path slightly thicker for visualisation purposes. We

have also adjusted Figure 1 with different colors to indicate three sub-groups (High Arctic, Terrestrial and Remote).

Line 155: please address the comment from Ashok Luhar; if the equations are not documented in previous work, please document them here (in particular how equations or parameters are altered for ozone deposition). It's unclear what COAREG is/does.

We have added Appendix B that includes the formulation of the air- and waterside resistance terms as well as the sensitivity to environmental factors.

Line 170: clarify whether only O₃ deposition follows COAREG in your simulations: what about other species (you say you are motivated to use this scheme because it provides consistency for all compounds)?

The scheme indeed allows for a similar and consistent representation of ocean-atmosphere exchange of other species. However, because of a lack of long-term and large-scale datasets (both in terms of input and validation) for other species a similar evaluation (as for O₃) is not possible. We have therefore decided to only include the representation of ocean-atmosphere of O₃. We have explicitly added this in the text.

Line 176: please explicitly say what MacDonald et al. 2014 does. Otherwise your reader does not know how to compare the MacDonald + Sherwen datasets

We have rewritten the sentences to make clear that MacDonald is a distribution that is solely dependent on SST.

Line 177: is there independent evidence that I_{aq} should be higher (and more like Sherwen) than MacDonald? Otherwise the authors need to say that this study assumes higher I_{aq} for the purposes of their investigation and that the I_{aq} values are highly uncertain (I hope this is something you plan to constrain in the upcoming field campaign)

In general, these I_{aq} distributions are highly uncertain for high latitudes due to the limited availability of observations. On the global scale, the Sherwen et al. (2019) distribution most accurately represents the observed I_{aq} (Sherwen et al. 2019). Therefore, we have chosen to use this distribution (see also additional information in the discussion). We have explicitly added a statement in the methods to motivate the choice for Sherwen et al. (2019).

Line 179: to my understanding, other studies do consider DOM, but they find the effect to be low. Please clarify this here and in the introduction. generally, the discussion of other compounds in seawater could be more consistent throughout the text. I didn't find the sensitivity analyses in the discussion necessary given the lack of details provided.

We agree that the mentioning of DOM is not appropriate here (in the methods section) and have removed it. In the introduction we have mentioned the role of DOM since it has been addressed in multiple earlier studies of which some find a significant role of DOM (e.g. Chang et al. (2004), Ganzeveld et al. (2009), Martino et al. (2012)). In the discussion we reflect on the potential sensitivity to DOM-O₃ and DMS-O₃ reactions since they were not included in this study. We have chosen to perform extra sensitivity analysis with the same reactions from Ganzeveld et al. (2009) that found a global sensitivity. However, in this study (for Arctic summer), the oceanic Chlorophyll and DMS concentrations are too low to make a significant contribution to the oceanic O₃ deposition flux. We find it important to at least discuss and consider these reactants as a potential (significant) enhancement of O₃ deposition (which is often ignored) and have therefore performed the sensitivity studies.

Line 181: this was recently summarized in Clifton et al. Reviews of Geophysics 2020. I think current understanding is that the POSITIVE fluxes are due to chemistry. I think you should clarify here, and perhaps include the range of observed ozone deposition velocities over snow that Clifton provides

We have added the reference to Clifton et al. (2020) including the observed range of O₃ deposition velocities.

Line 188: please clarify what happens in the WRF-Chem Wesely scheme. Is it exactly the Wesely scheme or some derivative?

For deposition to oceans and snow/ice this is exactly the Wesely scheme with a constant surface uptake resistance as described in Wesely (1989).

Line 195-205: references for diurnal cycle controls on ozone over high arctic vs. terrestrial vs. remote sites?

We have included references to studies that address the (lack of) controls of diurnal cycle in surface O₃.

Line 215: satellite observations of what?

CAMS assimilates satellite observations of O₃ that therefore mostly affects the stratospheric O₃ contribution. We have included the explicit mentioning of 'O₃' in the manuscript.

Line 230: I think it's important to show I_{aq} concentrations at least in the supplemental because this is an important assumption of your study, and it's not obvious to the reader where concentrations would be high vs. low

We have included the I_{aq} distribution in Appendix C and have referred to this while discussing the results to help the reader interpreting the results.

Line 231 - new paragraph should start at "Figure 3c shows..."

We have started this line with a new paragraph.

Line 231: This seems like the first time we are hearing about Ra aside from the very quick general definition. Maybe more introduction to this term in the intro is needed (i.e., what does it depend on?) Also, does Ra change from Wesely to COAREG?

Because O₃ deposition is especially restricted to the surface uptake resistance term we have focused on the description of that term (also indicated in the introduction). We have elaborated more on the r_a and r_b terms in the introduction but the main point that is that it depends on the efficiency turbulent transport to the surface, both in the COAREG and Wesely scheme. There are small differences in the definition of the r_a term but the model is not sensitive to this representation since the r_a term only becomes important at low wind speeds for both representations. The r_a term of COAREG is included in Appendix B to illustrate the role of e.g. the friction velocity.

Line 240: Have you isolated that temporal variability in ozone deposition velocity is +/-20% just due to waterside turbulent transport? Also, how much is the temporal variability in ozone deposition velocity in the default scheme? It could be 20% as well. So, saying variability from COAREG is +/-20% is not very compelling.

Because the surface resistance term in the COAREG scheme is up to 5x higher compared to the Wesely scheme the r_a term becomes even less important in the COAREG simulations. Since there is no short-term variability in other drivers of O₃ deposition in the model (e.g. SST and I_{aq}) the variability expresses the role of waterside turbulent transport.

Figure 3: First, I don't think this figure is colorblind friendly. Second, I understand that the authors want to use different colormaps because the ranges of the values are different, but the purple in both is confusing. What about just two different single-hue color bars? Third, is the point of (c) to show differences in variability or magnitude? I think the former, since the magnitude differences are shown by (a) and (b). Would recommend having two difference y-axes for default and COAREG on (c) so one can see differences in variability more. It would also be helpful to have windspeed on this plot, since the authors talk about changes in Vd with wind speed in the text.

We have changed the colormaps in Fig. 3a,b to colormaps that should be more colorblind friendly. Please let us know if this is not the case. The point of Fig. 3c is to show the typical variation within the simulation in V_{d,O_3} for both the NUDGED and COAREG runs. Based on comments by another reviewer we have added in Appendix B. (Fig. B1) the sensitivity to environmental factors such as wind speed. We have tried adding the simulated wind speeds in Fig. 3c but the Figure became quite messy. We hope that by including Fig. B1 the sensitivity and role of wind speed in Fig. 3c becomes clear.

Table 1: Why are there slightly differences in terrestrial Vd between default and COAREG? Please compare COAREG to the nudged simulation...

We have updated Figure 4 to include the NUDGED simulation instead of the DEFAULT simulation and have updated Table 1 to also give the results of NUDGED accordingly. Furthermore, we have changed the text in Sect. 3.1 to refer to the NUDGED run instead of the DEFAULT run. The results and conclusions drawn from Figure 4 and Table 1 are equal for the DEFAULT and NUDGED runs. To clarify, there are slight differences between COAREG and DEFAULT due to the different representation of meteorology affecting r_a , r_b and r_{stom} . Between COAREG and NUDGED less deposition to oceans would lead to higher O_3 over land in some instances that increases the total deposition budget.

Section 3.2: This section needs revising. It jumps around between talking about how COAREG changes things vs. spatial variability generally, and I'm not sure what I should be 'taking away'. What needs to be clear is how COAREG improves the simulation of monthly mean ozone spatial variability.

We have restructured this Section to first discuss the similarities and overall surface O_3 concentrations over land before addressing the role of the adjusted deposition scheme. We have also added a paragraph break to make this more clear for the reader. By also removing the comparison with CAMS here the structure of the Section should be more clear.

Line 252: The authors can be more definitive here. Also, what are the authors getting at? There are clear changes in Vd and the budget. . . Perhaps the authors mean that the differences across simulations are not reflected in the site-level monthly mean evaluation. Clarity needed.

The point is that even though the change in O_3 dry deposition is very limited in absolute terms, comparing this to the total O_3 deposition budget (Tab. 1), is has a large influence on the concentrations and distribution of O_3 over oceans and sea ice as illustrated in Fig. 4 once again indicating the need of these mechanistic representations in (other) ACTMs. We have better connected Sect. 3.1 and 3.2 by restructuring the Section (based on previous comment)

Line 254: This is not a complete sentence. Also, what am I supposed to contrast?

We have removed this sentence as it was misplaced in the results section.

Line 273-9: I'm not quite sure what we are learning from the CAMS reanalysis, and I find it particularly confusing to have it discussed in each section. In the least, I suggest all discussion of CAMS be moved to a separate section at the end. I don't know enough about CAMS to be able to interpret the meaning or cause of differences.

As also indicated in the main comments above we have removed the CAMS analysis for this section and have put less emphasis on CAMS in other sections but have left it in the manuscript as CAMS is an important product that is widely used to constrain atmospheric chemistry models e.g. the one used in this study but also to provide information about atmospheric composition/air quality in remote locations such as the High Arctic.

Figure 4: It really does not make sense to me to show the default simulation in Figure 4. The nudged simulation should be shown here to illustrate differences due to ozone deposition, the point of the paper. I think the authors need to present some statistics as to how the different model simulations capture spatial variability in monthly mean ozone.

We agree that it is better to compare the NUDGED and COAREG simulations here and have changed the figure accordingly. In this case, the results/conclusions by changing DEFAULT to NUDGED do not change since nudging the model to ERA5 mostly affects the temporal variability of O₃ and not the monthly mean concentrations.

Figure 5: I find it strange that the authors are just focusing on mean bias and MAE here, when they say in the text that they want to look at short term variability. How does COAREG improve variability? It would be helpful for the reader if the authors included some information as to how the diurnal cycle of Vd changes with COAREG. Is the important thing the diurnal cycle or day to day variability?

Another motivation of this study is to evaluate this mechanistic representation of O₃ ocean-atmosphere exchange with a large dataset of observed hourly surface O₃ concentrations at multiple (25 sites) which has, to the authors' knowledge, not been done before. To perform the evaluation we have chosen to show the evaluation for all sites to indicate for which areas the new modelling setup is most sensitive (in this case the High Arctic sites because of the deposition footprint). Thereafter, we go into more detail for a selection of the sites (6 High Arctic sites, Section 3.4) by showing the short-term variability at these individual sites. The introduction statement of this Section 3.3 might have been misplaced and has been revised. The short-term (days-weeks) variability in V_{d,O₃} to oceans is driven to a large extent by wind and therefore does not show a clear diurnal cycle in contrast to V_{d,O₃} to vegetation. The day-to-day variability in surface O₃ arises from changes in synoptic conditions (by affecting the the V_{d,O₃} to oceans and advection of O₃) and boundary layer mixing (entrainment). Therefore, we have also isolated the 'Terrestrial' sites in the analysis that show a clear diurnal cycle in observed surface O₃.

Line 311-3: 'to a lesser extent' than what? Generally, closing with this statement makes me question the authors' use of a regional model here. Is this the authors' intention? I suggest revising.

We have opted for this regional modelling setup to focus on the short-term variability compared to other (often global and monthly averaged) studies (e.g. Ganzeveld et al. (2009), Pound et al. (2019)). The domain setup has been selected in such a way that the simulated results, and especially those evaluated with observations, are as least as possible influenced by the boundary conditions (also considering computing costs etc.). However, a general implication of a regional modelling setup is that the simulated results near the edges of the domain (e.g. the observations in Scandinavia) are generally more influenced by advection over the edges of the domain from the CAMS product. This statement is indeed not appropriate in the Results section and does not add substantial information to this Section and has consequently been removed.

Section 3.4: This section could be more quantitative. It's unclear why the authors chose to discuss some features of the intercomparison and not others.

Also based on comments by Reviewer #1 we have rewritten this section to also include the standard deviation of observation minus prediction and have elaborated on some of the features of the intercomparison to put more emphasis on this short-term temporal variability in observed and simulated surface O₃ concentrations.

Line 318: How do the authors select the sites used in Figure 4? Do they just choose the ones at which that the COAREG scheme performs best? This is concerning, given that the title and conclusions majorly depend on Figure 4.

We assume 'Figure 4' is a typo and refers to Figure 6 since Sect. 3.4 and line 318 refer to Figure 6. The sites shown in Figure 6 are selected based on the criteria in Sect. 2.3. Namely, these are all the 'High Arctic' sites (all sites > 70 °N) and make up all the data presented in Fig. 5a-d. These sites have been selected having a deposition footprint being a combination of (sea-)ice and oceans because of their location. Sites in the 'Terrestrial' category generally do not benefit from the addition of the COAREG exchange routine as indicated in Sect. 3.2 and Sect. 3.3 (Figure 5). In the 'Remote' category we also find an improvement of model simulated surface O₃ (Figure 5). However, this is limited to individual sites (e.g. Lerwick, Storhofdi) that are close to the coast. We included in the results section again the reasoning behind the selection of these 'High Arctic' sites.

Line 327: new paragraph starting at 'At Summit,'

New paragraph started for all site descriptions.

Line 334: cut 'Interestingly' and start new paragraph here.

New paragraph started for all site descriptions.

Line 376-8: exactly why the nudged simulation should be the 'default' simulation here

We hope that the revised setup of the paper mainly focussing on the NUDGED and DEFAULT runs (by also changing Fig. 3, Table 1, Fig. 4, Table 2 and Fig. 6) better reflects the main objective of the manuscript.

Line 390: say why: because there are no observations, right?

We have added 'due to a lack of oceanic O₃ deposition measurements'. We have also removed the first sentence of this paragraph which included similar information.

Line 443: spelling error

Changed to 'meteorology'

Line 445: new paragraph here

We have removed multiple statements from the following paragraph to make it one (shorter) paragraph to reduce the already lengthy discussion.

Line 459-66: please cut this paragraph of 'next steps' in an already lengthy discussion; it's not really appropriate for a paper

Due to the already quite large Discussion Section we have removed this paragraph. Based on the comments by another reviewer we did include the need for additional observations to better constrain these modelling studies.

Line 470: for all trace gases or just for ozone here?

As indicated in one of the comments above, and now also included in the Methods Section, this is only included for O₃. We have therefore adjusted the statements in the Conclusion.

Line 490-3: this is similar to the finding of Clifton et al. 2020 10.1029/2020JD032398 that when the ozone lifetime is long ozone is very sensitive to small changes in a small deposition velocity

We have included references to Clifton et al. 2020 when we discuss the results on this sensitivity to small changes in deposition velocity in shallow ABLs

Line 505: why is this revision needed at the global scale? Why is short term variability in ozone at high arctic sites important for ozone globally?

With Arctic climate being relevant for global climate change and ozone being part of that climate change signal, better quantification and representation of O₃ including the role of deposition in the Arctic is one motivation for this study. The significance of evaluating the role of deposition in explaining short-term variability is that we show, especially by introducing the more mechanistic deposition representation that it significantly improves the skill of the model to capture in-situ O₃ concentration measurements, with these observations also being used to evaluate the performance of any ACTM on Arctic composition. It secures a more fair observation-model comparison. This manuscript also shows that the revised deposition scheme reduces the bias on longer timescales (monthly averages) which corroborates the findings of e.g. Pound et al. (2019). We are aware that we did not perform a global evaluation such a coupled modelling setup but opted for a regional approach to also address the short-term variability often not included in other studies. However, by including a large observational datasets (25 stations) this manuscript shows that the mechanistic ocean-atmosphere deposition approach and the changes to

sea-ice and snow deposition improves the comparison at those sites that have a deposition footprint that is mostly affected by these surface types.

Line 508: what is the 'fate of the arctic O3 budget'?

We have changed this to 'future trends in Arctic tropospheric O₃' to indicate the importance of an accurate representation of the deposition sink term in predictions of O₃ trends.

Review #3:

1. Influence of land deposition?

Near-surface ozone is a fascinating chemical compound, influenced by many, sometimes offsetting, processes. Models may get right concentrations for some wrong reasons or get it wrong for the right reasons. This study focused on late summer (August to early September) when deposition over land vegetation can still play an important role in influencing surface ozone concentrations at observational sites in northern high-latitudes (e.g., sites in Norway, Sweden, and Finland). Throughout the results section in this paper, however, there are no discussion on the potential influence of land deposition processes. There are studies showing that changes in dry deposition schemes over land can lead to as much as 10 ppbv differences in simulated mean surface ozone concentrations at northern high-latitude sites. Please see Figures 13 to 16 in Lin et al. (GBC 2019) and discussions therein. Although the present study focused on oceanic deposition, the potential influence of land deposition needs to be discussed.

We thank reviewer #3 for this review which is mostly addressing the role of O₃ deposition to land and vegetation. In this study we focused on the role of O₃ deposition to oceans and sea ice and therefore found most significant results at locations with a close to the sea-ice and oceans. We are very much aware that some of the modelled surface O₃ concentrations at sites in e.g. Norway, Sweden and Finland (mostly in the 'Terrestrial' group) are not always represented accurately illustrated by Fig 5. i-l. This Figure indicates that, at least in terms of magnitude of monthly mean O₃, the model performs quite well (low Bias). However, this Figure also indicates quite some spread around the mean indicated by a Mean Absolute Error which is similar to some of the MAE's we found at 'High Arctic' and 'Remote' sites. We expect that land deposition, but also other factors such as emissions of precursors (from biogenic and anthropogenic sources) and the diurnal cycle in boundary layer mixing, will play an important role at the Terrestrial sites which are located more inland compared to the sites from High Arctic and Remote groups. We have added a section in the Discussion to discuss the potential role of land deposition on the results we found in our study, especially related to the Terrestrial sites. We have also compared the simulated O₃ deposition velocities to vegetation and land to observational studies (e.g. van Dam et al. (2016)).

2. Chemical boundary conditions?

It is not clear from Section 2 whether the WRF-Chem simulations use chemical boundary conditions from a global model, which can potentially influence near-surface ozone concentrations at remote Arctic sites.

We have used the ERA5 (meteorology) and the CAMS (chemistry) products as initial and boundary conditions as indicated in Section 2. We have explicitly added the boundary conditions used also in Table A1.

3. Fig.4d: Need to include comparisons of ozone frequency distributions with observations, at least at sites where measurements are available. Justification to compare with CAMS reanalysis product is not clear. CAMS products are NOT observations.

Also based on comments of reviewer #2 we have reduced Fig. 4 back to two panels showing the spatial distribution of monthly mean surface O₃ of (a) NUDGED and (b) COAREG to illustrate the effect of the revised deposition scheme on the long-term averaged surface concentrations. Therefore, we have also removed CAMS in this comparison to avoid readers interpreting CAMS as observations. And we are indeed aware that the CAMS data can not be interpreted as observations but with this reanalysis product being as much constrained as possible with observations is comes as a large-scale data-assimilation probably as close as possible to those observations.

4. Fig.5 and Fig.6: The referee suggests removing results from the DEFAULT simulation without nudging when comparing hourly ozone with observations. We all know that the DEFAULT simulation without nudging is not expected to simulate the synoptic day-to-day variability of ozone in observations. Including DEFAULT makes the plots (e.g., Fig.6) messy and makes it difficult for readers to see the impact of interactive ocean deposition.

We have removed the DEFAULT simulation from Fig. 6 to make this Figure and Section focus more on the impact of interactive ocean deposition on simulated hourly O₃ concentrations. We have also made changes to the text accordingly.

5. Label the site names shown in Fig.6 on the maps in Fig.4 to facilitate understanding. Separate analysis for coastal versus far-inland sites can be a way to illustrate the influence of oceanic versus land deposition.

We have tried to include the stations shown in Fig. 6 to Fig. 4. However, the figure got quite messy and hard to interpret. Instead, we have updated Fig. 1 (model domain) with different colours to differentiate between the three sub-groups: High-Arctic, Remote and Terrestrial. Furthermore, the label for Zeppelin was missing and was added.

6. Label correlations and mean biases for each model, directly in Fig.6 (not in table), to facilitate understanding.

We have tried to include the include the correlations and bias for the model simulations to Fig. 6. However, including three metrics (after also including standard deviation of Observations-Prediction based on the comments by Reviewer #1) for the three simulations (NUDGED, COAREG and CAMS) made the Figure quite messy. By removing DEFAULT from Fig. 6, and rewriting some of the text of Section 3.4 also based on comments of other reviewers we hope to facilitate better understanding of Figure 6 and Section 3.4.

7. References in Introduction need to be updated to include more recent findings. For example,

Line 35-40, Ozone sources and sinks

Added references to Young et al. (2018) and Tarasick et al. (2019)

Line 42: The role of emission changes on mid-latitude ozone trends

Added reference to Lin et al. (2017)

Lines 47-50: Dry deposition processes over land and the importance of interactive ozone deposition on surface ozone variability

Added references to Kavassalis & Murphy (2017), Lin et al. (2019) and Lin et al. (2020)

Short comment #1:

*Hello, I just wanted to let you know about my recent paper on long-term ozone trends across the globe. We looked at trends at 6 Arctic sites with data through the year 2017 or 2018 (Barrow, Alert, Denali, Zeppelin, Esrange Tustavartn). We report trends for the full records and since the year 2000. Table 2 in the main text and Figure S-3 in the Supplement contain the numbers with most relevance to your study. Also, Appendix S-B in the Supplement has trend plots for the individual sites. I hope you find these results useful when you mention long-term trends in the Introduction. Best regards Owen Cooper
University of Colorado Boulder/NOAA CSL*

We thank Owen Cooper for informing us about this excellent recent paper on long-term O₃ trends at remote sites. Unfortunately, we missed the release of your paper while writing this manuscript. We have updated the introduction with references to Cooper et al. (2020) where relevant.

Short comment #2:

Thank you to the authors for presenting a very interesting study.

I would like to highlight one aspect of the paper where there appears to be an ambiguity. The authors have coupled the Coupled Ocean-Atmosphere Response Experiment Gas transfer algorithm (COAREG, version 3.6) to the regional WRF-Chem model. This model setup supposedly includes an improved (two-layer?) mechanistic scheme for the calculation of the waterside surface resistance term in computing ozone dry deposition to water, but the authors have not presented any equations/parameterisations that have been used for this term. They refer to the paper by Porter et al. (2020) for COAREG (version 3.6) and looking up this paper I do not see any application to ozone deposition there (only water vapor and sulphur dioxide are considered). Other papers are also cited but I do not think they relate to version 3.6.

Therefore, it is not clear what exact equations for the parameterisation of the waterside surface resistance term (and associated parameters such as iodide concentration in water, reaction rate constant and ozone solubility) for ozone deposition have been used, and there does not appear to be a source for finding these. It is will be useful for the authors to present these equations in the paper for the sake of completeness and clarity.

We thank Ashok Luhar for his kind words and addressing the ambiguity regarding the details of the COAREG exchange routine. First of all, we have removed the reference to Porter et al. (2020) in Sect. 2.2 to avoid ambiguity since this paper does indeed not address deposition of O₃. The version of COAREG used in this study is the version in Fairall et al. (2007, 2011) extended with a two-layer scheme based on Luhar et al. (2018). We have added the formulation and formulas of the simulated deposition velocities including the air and waterside resistance terms in Appendix B. This includes the definition of the associated parameters such as the solubility of O₃, chemical reactivity and molecular diffusivity. In our manuscript we use the Iodide distribution from Sherwen et al. (2019) which we have once again mentioned in Appendix B.

Since there is no specific manuscript available that describes this version of COAREG we hope that we have removed the ambiguity by adding Appendix B and by a now more connected description in the main text: "Here we use COAREG version 3.6, which is extended with a two-layer scheme for surface resistance compared to the previous version described by Fairall et al. (2007, 2011). The two-layer scheme is similar to the work by Luhar et al. (2018) building upon ..."

Role of oceanic ozone deposition in explaining short-term variability of Arctic surface ozone at high-Arctic sites

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Abstract. Dry deposition is an important removal mechanism for tropospheric ozone (O₃). Currently, O₃ deposition to oceans in atmospheric chemistry and transport models (ACTMs) is generally represented using constant surface uptake resistances. This is despite the fact that considering the role of solubility, waterside turbulence and O₃ reacting with ocean water reactants such as iodide ~~and dissolved organic matter~~ results in substantial spatiotemporal variability in O₃ deposition and concentrations in marine boundary layers. We hypothesize that O₃ deposition to the ~~cold~~-Arctic ocean, ~~with having a~~ relatively low reactivity, is ~~also~~ overestimated in current models with consequences for ~~background tropospheric~~ concentrations, lifetime of O₃ and long-range transport of O₃. In this study, we investigate the ~~role-impact~~ of the representation of oceanic O₃ deposition to the simulated magnitude and spatiotemporal variability in Arctic surface O₃. ~~This study also serves as a preparatory study to understand the year-round Arctic O₃ concentration and deposition flux measurements as part of the MOSAiC field campaign. Furthermore, it is also important to enhance our understanding and quantification of Arctic ocean-atmosphere exchange of O₃ and other climate-active trace gases given the anticipated opening of the Arctic ocean.~~

We have ~~coupled-integrated~~ the Coupled Ocean-Atmosphere Response Experiment Gas transfer algorithm (COAREG) ~~to-into~~ the mesoscale meteorology and atmospheric chemistry model Polar-WRF-Chem (WRF) ~~and introduced-which introduces~~ a dependence of O₃ deposition on ~~ocean-waterside turbulent mixing conditions and biogeochemical composition~~ physical and biogeochemical drivers of oceanic O₃ deposition. We have also reduced the O₃ deposition to sea ice and snow. Here, we evaluate the performance of WRF and the CAMS reanalysis data against hourly-averaged surface O₃ observations at 25 sites (latitudes > 60 °N) including the ~~ASCOS~~ Arctic Summer Cloud Ocean Study (ASCOS) campaign observations. This is the first time such a coupled modelling system has been evaluated against hourly observations at Pan-Arctic sites to study the sensitivity of the deposition scheme to the magnitude and short-term temporal variability in Arctic surface O₃. We also analyze the impact of nudging WRF to the synoptic conditions from the ECMWF ERA5 reanalysis data on simulated Arctic meteorology and comparison of observed and simulated O₃ concentrations.

We show that the more mechanistic representation of O₃ deposition over oceans and reduced snow/ice deposition improves simulated Arctic O₃ mixing ratios both in terms of magnitude but also regarding observed temporal variability. Using the newly implemented approach, O₃ deposition velocities have been simulated in the order of 0.01 cm s⁻¹ compared to ~0.05 cm s⁻¹ in the constant surface uptake resistance approach. The simulated monthly-mean spatial variability in the mechanistic approach (0.01 to 0.018 cm s⁻¹) expresses the sensitivity to chemical enhancement with dissolved iodide whereas the temporal variability

(up to $\pm 20\%$ around the mean) expresses mainly differences in waterside turbulent transport. The bias for all observational sites above 70°N reduced from -7.7 ppb to 0.3 ppb with nudging and the revision to ocean and snow/ice deposition. Our study confirms that O_3 deposition to high-latitude oceans and snow/ice is ~~overestimated in current models~~ generally overestimated in ACTMs. We recommend that a mechanistic representation of oceanic O_3 deposition ~~is~~ should be used in ACTMs to improve the representation of Arctic surface O_3 concentrations in terms of magnitude and short-term temporal variability. ~~The revised ocean-atmosphere exchange representation can be further refined using the MOSAiC flux measurements as well as complementary observations such as sea ice and ocean water iodide concentrations.~~

1 Introduction

35 Tropospheric Ozone (O_3) is the third most important greenhouse gas and a secondary air pollutant negatively affecting human health (Nuvolone et al., 2018) ~~and~~ plant growth (Ainsworth et al., 2012) and artificial materials such as rubber (Lee et al., 1996) due to its oxidative character. O_3 shows a large spatiotemporal variability due to its relatively short lifetime (3-4 weeks) in the free troposphere compared to other greenhouse gases. Its main sources are chemical production and entrainment from the stratosphere. Its main sinks are chemical destruction and deposition to the Earth's surface (Young et al., 2018; Tarasick et al., 2019).
40 . Understanding the Arctic O_3 budget is of particular interest because its remote location implies that anthropogenic sources and sinks are generally absent. This makes these ~~background-Arctic~~ O_3 observations ~~an excellent indicator for global trends (Helmig et al., 2007b; Gaudel et al., 2020)~~ excellent indicators for global trend analysis (Helmig et al., 2007b; Gaudel et al., 2020; Cooper et al., 2014). In the Arctic, routine tropospheric O_3 observations indicate an increasing trend up to the early 2000s which is leveling off (Oltmans et al., 2013; Cooper et al., 2014) ~~or decreasing at individual sites (Cooper et al., 2020)~~ in the last decade (Oltmans et al., 2013; Cooper et al., 2014).
45 . This upward trend can be attributed to increased emissions of precursors in the mid-latitudes (~~Cooper et al., 2014~~) ~~but also (Cooper et al., 2014; Lin et al., 2017), but also changes in O_3 deposition to vegetation as a result of droughts and heatwaves (Lin et al., 2020) and~~ stratosphere-to-troposphere transport may have played a role (Pausata et al., 2012). Local emissions of precursors are expected to become an important source of Arctic O_3 concentrations due to the warming Arctic climate and increasing local economic activity (Marelle et al., 2016; Law et al., 2017). This ~~stresses the need to better understand~~ underlines the need for understanding the sources and sinks of Arctic tropospheric O_3 and to accurately represent them in atmospheric chemistry and transport models (ACTMs).

On the global scale, dry deposition accounts for $\sim 25\%$ of the total sink term (Lelieveld and Dentener, 2000) in ACTM simulations and is especially important for the O_3 budget in the Atmospheric Boundary Layer (ABL) because it occurs at the Earth's surface (~~Hardaere et al., 2015~~) (Kavassalis and Murphy, 2017; Lin et al., 2019, 2020). Dry deposition in ~~such model assessments~~ ACTMs is often represented as a resistance in series approach (Wesely, 1989). In this approach the total resistance r_t is the sum of three serial resistances: the aerodynamic resistance (r_a) representing turbulent transport to the surface, the quasi-laminar sub layer resistance (r_b) representing diffusion close to the surface and the surface resistance (r_s) expressing the efficiency of removal by the surface. The dry deposition velocity (V_d) is then evaluated as the reciprocal of r_t . The r_a term is independent of the chemical species and mainly depends on the stability of the atmosphere and friction velocity

60 (u_*) (Padro, 1996; Toyota et al., 2016). The r_b term also scales with u_* and varies with the diffusivity of the chemical species (Wesely and Hicks, 2000). For very soluble species or reactive species such as nitric acid uptake by the ocean water is very fast, expressed by a (i.e. r_s of $\sim 0 \text{ s m}^{-1}$), implying that the other resistances determine r_t and thus V_d . Less soluble gases, like O_3 , have a high r_s that mainly, in comparison to the relatively small $r_a + r_b$ term, that dominates the magnitude of the O_3 dry deposition velocity (V_{d,O_3}). Thus, accurately representing the surface uptake efficiency is of high importance of O_3 is crucial.

65 Even though Observed O_3 deposition to oceans (e.g. Chang et al., 2004; Clifford et al., 2008; Helmig et al., 2012) and coastal waters (e.g. Gallagher et al., 2001) is relatively slow compared to terrestrial surfaces, expressed by typically observed ocean V_{d,O_3} of ($\sim 0.01\text{-}0.1 \text{ cm s}^{-1}$ (e.g. Helmig et al., 2012)), especially compared to observed maximum V_{d,O_3} for forests up to 2 cm s^{-1} (Fan et al., 1990). However, it plays a large role in the total O_3 deposition budget due to the large surface area of water bodies (Ganzeveld et al., 2009)(Ganzeveld et al., 2009; Hardacre et al., 2015). Recent experimental and modelling studies

70 indicate the spatiotemporal variability in oceanic O_3 uptake efficiency (Ganzeveld et al., 2009; Helmig et al., 2012; Luhar et al., 2018). However, most models-ACTMs often still use a constant O_3 surface uptake efficiency of 2000 cm s^{-1} to water bodies, proposed by Wesely (1989), resulting in a simulated ocean V_{d,O_3} of $\sim 0.05 \text{ cm s}^{-1}$. The observed V_{d,O_3} shows a larger variability including also a dependency on wind speed and Sea Surface Temperature (SST) (Helmig et al., 2012). The dependency on wind speed also expresses an enhancement of O_3 deposition due to waterside turbulence (Fairall et al., 2007).

75 This turbulence driven enhancement is complemented by a strong chemical enhancement of oceanic O_3 deposition associated with its chemical destruction through oxidation of ocean water reactants such as dissolved iodide and dissolved organic matter (DOM) (Chang et al., 2004). Mechanistic O_3 deposition representations in models include the physical and biogeochemical processes related to the exchange and destruction of O_3 in surface waters (Fairall et al., 2007, 2011; Ganzeveld et al., 2009; Luhar et al., 2017, 2018). Dissolved iodide is deemed to be the main reactant of O_3 in surface waters (Chang et al., 2004)

80 and therefore often applied in these representations. Some studies only consider dissolved iodide as a reactant (Luhar et al., 2017; Pound et al., 2019) whereas Ganzeveld et al. (2009) also included DOM as one reactant contributing to the chemical enhancement of oceanic O_3 deposition. However, the role of DOM in oceanic O_3 deposition remains difficult to quantify and which appears to be mainly addressed by controlled laboratory measurements-experiments or O_3 flux measurements at sites with elevated DOM water concentrations. Nevertheless, application of these more mechanistic ocean O_3 deposition representations

85 illustrated the importance of a more explicit representation of O_3 dry deposition in ACTMs, not only regarding the impact on marine ABL O_3 concentrations and budget, but also to consider potentially important feedback mechanisms. For instance, the implementation of these mechanistic exchange methods in ACTMs indicates a $\sim 50\%$ reduction of the global mean V_{d,O_3} which affects the tropospheric O_3 burden (Pound et al., 2019). This mechanistic representation especially results in a simulated decrease in V_{d,O_3} to cold polar waters with relatively low reactivity. Simulated V_{d,O_3} can be as low as 0.01 cm s^{-1} compared to

90 the commonly applied V_{d,O_3} of 0.05 cm s^{-1} in the constant surface uptake resistance approach (Pound et al., 2019). Regarding feedback mechanisms, consideration of the mechanisms that ultimately determine the efficiency of uptake and destruction of O_3 in ocean surface waters might also explain the release of halogen compounds into the ABL (Prados Roman et al., 2015). These halogen compounds-halogens, in turn, are involved in O_3 depletion in the ABL and therefore reduce further uptake and destruction of O_3 in ocean surface waters implying existence of a negative feedback mechanism.

95 Up until now, earlier studies ~~have mostly focused on the effects on the global scale (Ganzeveld et al., 2009; Luhar et al., 2017)~~
~~using on global scale oceanic O₃ deposition (Ganzeveld et al., 2009; Luhar et al., 2017)~~ mainly relied on the evaluation of
monthly mean surface O₃ observations (Pound et al., 2019). ~~However, the hypothesized reduction in O₃ deposition~~ The
implementation of these mechanistic exchange methods in ACTMs, in particular the method proposed by Luhar et al. (2018)
using a two-layer model representation (compared to a bulk layer version by Ganzeveld et al. (2009)), results in a ~50%
100 reduction of the global mean V_{d,O_3} which affects the tropospheric O₃ burden (Pound et al., 2019). The mechanistic representation
in Pound et al. (2019) especially results in a simulated decrease in V_{d,O_3} to cold polar waters with relatively low reactivity.
Simulated V_{d,O_3} can be as low as 0.01 cm s⁻¹ compared to the commonly applied V_{d,O_3} of 0.05 cm s⁻¹ in the constant surface
uptake resistance approach (Pound et al., 2019). However, the hypothesized deposition reduction to cold waters is ~~also~~ ex-
pected to substantially affect Arctic ABL O₃ concentrations on shorter timescales and potentially improve operational Arctic
105 O₃ forecasts, e.g. the air quality forecasts by the Copernicus Atmosphere Monitoring Service (CAMS) (Inness et al., 2019). ~~An~~
~~improved representation of sub-monthly Arctic O₃ concentrations helps to constrain the background O₃ concentrations in terms~~
~~of magnitude and variability whereas the~~
The evaluation of simulated oceanic O₃ deposition in the Arctic is hampered by a lack of O₃ ocean-atmosphere flux observa-
tions. Hence, evaluation of simulated O₃ deposition relies on evaluation of surface O₃ concentrations ~~not only regarding the~~
110 ~~simulated and observed magnitude but~~ in particular on the highly resolved temporal variability. We hypothesize that on the
daily and diurnal timescales these concentrations are largely controlled by temporal variability in the main physical drivers
of oceanic O₃ deposition, e.g. atmospheric and waterside turbulence. Chemical enhancement of, e.g., iodide to O₃ deposi-
tion is anticipated to control more the long-term (weeks-months) baseline level of V_{d,O_3} associated with anticipated ~~more~~
long-term (e.g. seasonal) changes in ocean water biogeochemical conditions (Sherwen et al., 2019). This evaluation of Arctic
115 spatiotemporal O₃ concentrations ~~in terms of magnitude and short- and long-term variability~~ aims to better understand sinks,
processes, feedbacks and impacts of Arctic air pollution (Arnold et al., 2016) and the role of long-range transport (e.g. Thomas
et al., 2013; Marelle et al., 2018) versus local sources (e.g. Marelle et al., 2016; Law et al., 2017; Schmale et al., 2018). Fur-
thermore, the ~~anticipated projected~~ opening of the Arctic ocean, as ~~one of the key features of Arctic~~ a result of climate change,
urges to improve our understanding of Arctic ocean-atmosphere exchange. ~~In this study we only focus~~ This study focuses on
120 the ocean-atmosphere exchange of O₃, but follow-up studies are planned with a focus on ocean-atmosphere exchange and ABL
concentrations of other trace gases such as dimethylsulfide (DMS), which enhances cloud formation and is involved in many
feedback mechanisms (Mahmood et al., 2019).
~~The main objective of this study is to address the role~~ We aim to identify and quantify the impact of a mechanistic representa-
tion of O₃ deposition in explaining observed hourly Arctic surface O₃ concentrations, both in terms of magnitude and temporal
125 variability. A mesoscale coupled meteorology-atmospheric chemistry model is ~~set up for an end-of-summer period in 2008 and~~
~~evaluated against a large dataset of pan-Arctic O₃ observations at a high resolution (hourly) timescale~~ for the end-of-summer
2008. Having a much higher spatial and temporal resolutions compared to other global modelling studies we aim to better
capture the role of spatiotemporal variability in O₃ deposition in explaining observed surface O₃ concentrations ~~in particular~~
particularly regarding temporal variability. We also indicate the role of meteorology in simulating these O₃ concentrations by

130 nudging the simulated synoptic conditions towards an atmospheric reanalysis dataset. ~~This study also serves as a preparatory study to understand the year-round Arctic O₃ concentration and deposition flux measurements including the role of the local meteorology such as boundary layer mixing and entrainment as part of the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAIC) campaign (-, last access: 16 September 2020).~~ Section 2 describes the adjustments to the deposition scheme [in the mesoscale ACTM](#), further model setup and observational datasets. Section 3 presents the main results of the
135 study which are further discussed in Sect. 4. This manuscript is finalized with the conclusions in Sect. 5.

2 Methods

2.1 Regional coupled meteorology-chemistry model

We use the Weather Research and Forecasting model (v4.1.1) coupled to chemistry (Chem) (Grell et al., 2005) and optimized for Polar regions (Hines and Bromwich, 2008). Polar-WRF-Chem (hereafter: WRF) is a non-hydrostatic mesoscale
140 numerical weather prediction and atmospheric chemistry model used for operational and research purposes. Figure 1 shows the selected study area including the locations of surface O₃ observational sites ~~that will be used in this study - selected for this study (more information in Sect. 2.3).~~ WRF is set up with a polar projection centered at 90°N, 250×250 horizontal grid points (30×30 km resolution) and 44 vertical levels up to 100 hPa, with a finer vertical grid spacing in the ABL and lower troposphere. The simulation period is ~~08-08-2008 to 07-09-2008~~ [08-August-2008 to 07-September-2008](#) including three
145 days of spin-up. This end-of-summer 2008 period is chosen ~~for two reasons:~~ 1) to limit the role of [active](#) halogen chemistry during springtime ([Pratt et al., 2013; Thompson et al., 2017](#)) ([Pratt et al., 2013; Thompson et al., 2017; Yang et al., 2020](#)) and 2) the additional availability of O₃ observations in the high Arctic over sea ice from the ASCOS campaign (Paatero et al., 2009). The ECMWF ERA5 meteorology (0.25°×0.25°) (Hersbach et al., 2020) and CAMS reanalysis chemistry (0.75°×0.75°) (Inness et al., 2019) products are used for the initial and boundary conditions. Boundary conditions, SSTs and sea ice fractions
150 are updated every three hours to these reanalysis products to allow for the sea ice retreat during the simulation. Other relevant parameterization schemes and emission datasets have been listed in Tab. A1 and are mostly based on Bromwich et al. (2013).

2.1.1 Nudging to ECMWF ERA5

The first WRF simulation, without any adjustments to O₃ deposition, indicated that WRF was misrepresenting the temporal variability in surface O₃ observations, most prominently starting from a few days into the simulation. We hypothesize that ~~these~~
155 ~~deviations are this misrepresentation is~~ caused by deviations in the synoptic conditions in the free running WRF simulation. ~~To verify this~~ [Hence](#), WRF results are compared against the observations from the Advanced Microwave Scanning Radiometer - Earth Observing System (AMSR-E) sensor on NASA's Aqua satellite. The near surface wind speeds above oceans from the Daily Level-3 data product are used with a spatial resolution of 0.25°×0.25° (Wentz and Meissner, 2004).

Figure 2 shows the temporal evolution in the bias (WRF minus AMSR-E) and Mean Absolute Error (MAE) of the daily and
160 ocean grid box averaged 10-m wind speeds. ~~The~~ [Although the](#) first days there is no clear bias. ~~However~~, later in the simulation

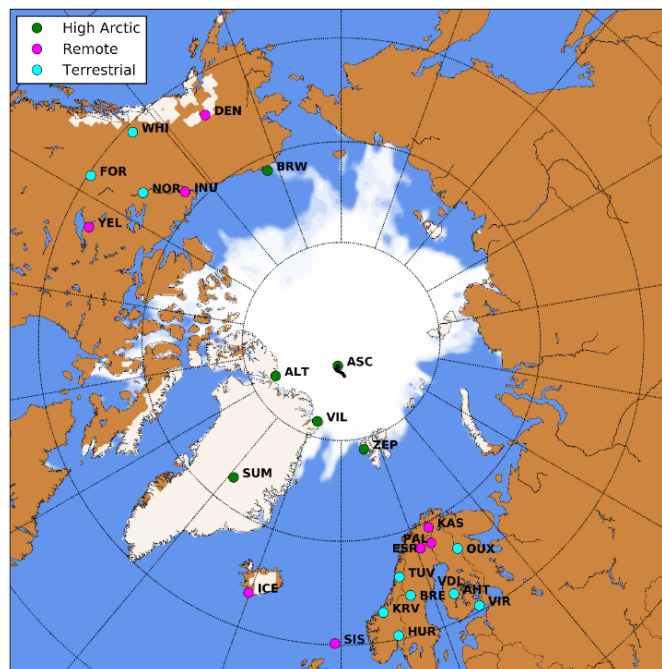


Figure 1. WRF domain including sea ice and snow cover at the start of the simulation. Locations with surface observations O_3 are indicated in green (High Arctic), magenta (Remote) and cyan (Terrestrial) (see Sect. 2.3). The drifting path of the ASCOS campaign during the simulation is indicated with the black line.

we find a persistent positive wind speed bias indicating that WRF overestimates the wind speeds above the Arctic ocean. During the first days the MAE amounts to $\sim 1.5 \text{ m s}^{-1}$, while later in the simulation the MAE reaches $2.5\text{-}3.0 \text{ m s}^{-1}$. To overcome the impact of this deficiency on our O_3 budget study, nudging is applied to ensure a fair model evaluation with observations. Hence, WRF is nudged every three hours to the ECMWF ERA5 humidity, temperature and wind fields in the free troposphere with nudging coefficients of $1 \cdot 10^{-5} \text{ s}^{-1}$, $3 \cdot 10^{-4} \text{ s}^{-1}$ and $3 \cdot 10^{-4} \text{ s}^{-1}$, respectively. In Sect. 3.3 the role-impact of nudging on simulated surface O_3 is further analysed.

2.2 Representation of ocean-atmosphere gas exchange

The Coupled Ocean-Atmosphere Response Experiment (COARE) (Fairall et al., 1996) has been developed to study physical exchange processes (sensible heat, latent heat and momentum) at the ocean-atmosphere interface. Later, COARE has been extended to include the exchange of gaseous species such as O_3 , dimethyl sulfide (DMS) and carbon dioxide (CO_2) (Fairall et al., 2011). Many studies have used the COARE Gas transfer algorithm (COAREG) in combination with eddy covariance measurements to study the effects of wind speed and sea state on ocean-atmosphere gas exchange (e.g. Helmig et al. (2012), Blomquist et al. (2017), Bell et al. (2017), Porter et al. (2020)). Furthermore, the COAREG algorithm has also been previously used in global O_3 modelling studies Ganzeveld et al. (2009). The choice for COAREG as ocean-atmosphere exchange param-

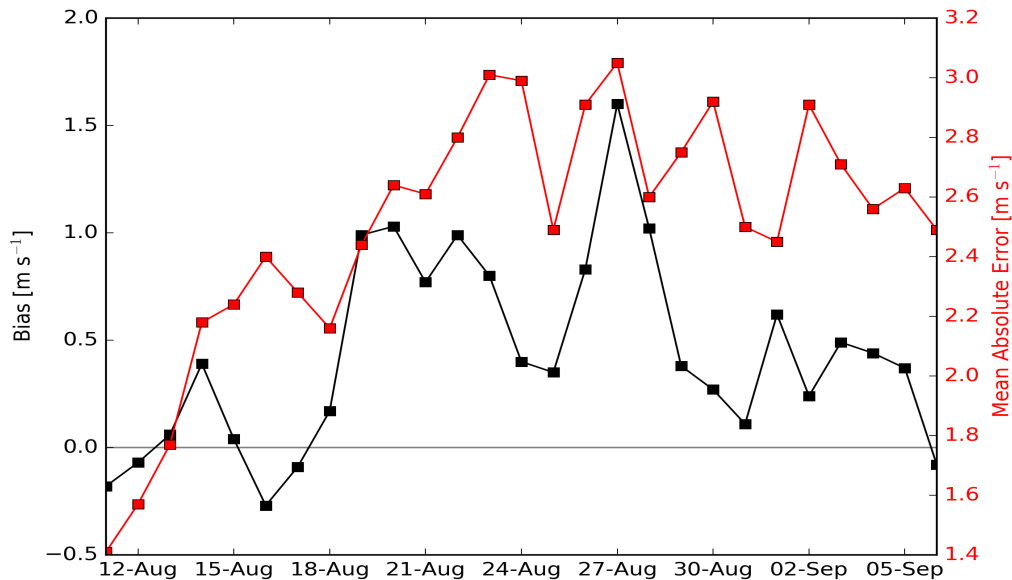


Figure 2. Temporal evolution of the bias (WRF minus AMSR-E, black) [m s^{-1}] and Mean Absolute Error (MAE, red) [m s^{-1}] of 10-m wind speeds above oceans for the period of 11-Aug to 6-Sep 2008. Note that the right y-axis starts at 1.4 m s^{-1} .

175 eterization is further motivated by the consistent coupling with other species such as DMS.
 Here we use COAREG version 3.6, which is extended with a two-layer scheme for surface resistance compared to the previous
 version described by Fairall et al. (2007, 2011). ~~COAREG version 3.6 has already been used in a study by Porter et al. (2020)~~
~~on air-sea transfer of highly soluble species.~~ The two-layer scheme is similar to ~~the work by~~ Luhar et al. (2018) building upon
 a first application of a 1-layer version of COAREG ~~for oceanic O_3 deposition in a global modelling study~~ by Ganzeveld et al.
 180 (2009). In that study, chemical enhancement of ocean O_3 deposition by its reaction with iodide was considered using a global
 climatology of ocean surface water concentrations of nitrate serving as a proxy for oceanic iodide concentrations (Γ_{aq}), the
 compound that is generally deemed to be the most significant reactant for O_3 in ocean water (Chang et al., 2004). Besides
nitrate, satellite-derived chlorophyll- α concentrations have been used as a proxy for Γ_{aq} (Oh et al., 2008). Since then, alterna-
 tive parameterizations of oceanic Γ_{aq} have been proposed (e.g. MacDonald et al., 2014) using SST as a proxy for this reactant.
 185 In COAREG, chemical reactivity of O_3 with Γ_{aq} is present through the depth of the oceanic mixing layer. O_3 loss by waterside
 turbulent transfer is negligible in the top water layer (few micrometers), but is accounted for in the underlying water column.
 The waterside turbulent transfer term is especially relevant for relatively cold waters because the chemical enhancement term is
 then relatively low (Fairall et al., 2007; Ganzeveld et al., 2009; Luhar et al., 2017). The last two important waterside processes
 that determine the total O_3 deposition are molecular diffusion and solubility of O_3 in seawater which both depend on the SST.
 190 In Appendix B we list the formulation of the air- and waterside resistance terms in the COAREG routine applied in this study

and show the sensitivity to the environmental factors wind speed, SST and Γ_{aq} for typical Arctic conditions.

In this study, the COAREG algorithm is coupled such that WRF provides the meteorological and SST input for the COAREG routine. In turn, the COAREG calculated ocean-atmosphere exchange velocities are used in the WRF model to calculate the ~~total flux. This study focuses on the exchange, in this case deposition, of O_3 . The oceanic O_3 deposition fluxes replace the default flux replacing the default oceanic O_3 deposition fluxes calculated by the Wesely (1989) scheme reflecting use of the default constant r_s of 2000 s m^{-1} .~~ For grid boxes with fractional sea ice cover, COAREG replaces the Wesely deposition scheme for the fraction that is ice free. Note that in this study, only O_3 ocean-atmosphere exchange is represented by COAREG not having modified simulations of ocean-atmosphere exchange of other compounds (e.g. DMS).

Moreover, we apply the monthly-mean Γ_{aq} distribution by Sherwen et al. (2019) ($0.125^\circ \times 0.125^\circ$ resolution). ~~This distribution does not only depend on SST, but which~~ applies a machine learning approach, namely the Random Forest Regressor algorithm (Pedregosa et al., 2011), using various physical and chemical variables ~~For such as SST, nitrate and chlorophyll- α .~~ This distribution replaces the previously applied Γ_{aq} estimations only using SST (Chance et al., 2014; MacDonald et al., 2014). At high latitudes, ~~this implies higher these Γ_{aq} and thus higher~~ distributions are highly uncertain due to the limited number of observations. However, the choice for Sherwen et al. (2019) is motivated by the most accurate representation of observed Γ_{aq} on the global scale. Figure C1 shows the spatial distribution of Γ_{aq} used in the calculation of the O_3 deposition ~~compared to MacDonald et al. (2014). In that study~~ velocities of COAREG coupled to the WRF model. Using the Sherwen et al. (2019) distribution for August/September we found relatively high Γ_{aq} concentrations ranging between 30 nM and 80 nM for the open oceans up to 130 nM in coastal waters. In MacDonald et al. (2014) and Chance et al. (2014), Γ_{aq} is solely a function of SST which leads to ~~very small Γ_{aq} in the cold Arctic ocean order of 5 to 50 nM~~ and thus low reactivity and O_3 deposition velocities. ~~As mentioned previously, the study by Ganzeveld et al. (2009) also considered the potentially important enhancement in oceanic O_3 deposition by its reaction with DOM, a feature not considered in studies by Luhar et al. (2017); Pound et al. (2019). In Sect. 4 we will discuss the potential role of DOM in our simulations and Arctic O_3 deposition.~~

2.2.1 Deposition to snow and ice

Reported atmosphere-snow gas exchange spans a wide range of observed O_3 deposition velocities. Some studies even report episodes of negative deposition fluxes (emissions) over snow or sea ice (Zeller, 2000; Helmig et al., 2009; Muller et al., 2012). Clifton et al. (2020b) recently summarized observed O_3 deposition velocities to snow having a range of -3.6 to 1.8 cm s^{-1} with most of the observations indicating a deposition velocity between 0 and 0.1 cm s^{-1} for multiple snow covered surfaces (e.g. grass/forest/sea-ice). Generally, ozone concentrations in the interstitial air of the snowpack is lower than in the air above making it a not a direct source of O_3 in terms of emissions (Clifton et al., 2020b). However, the emissions of O_3 precursors from the snowpack can enhance O_3 production in the very stable atmosphere above the snowpack (Clifton et al., 2020b). Helmig et al. (2007a) investigated the sensitivity of a chemistry and tracer transport model to the prescribed O_3 deposition velocity and found best agreement between modelled and observed O_3 concentrations by applying deposition velocities in the order of 0.00 - 0.01 cm s^{-1} . Based on Following Helmig et al. (2007a) we have increased the O_3 surface uptake resistance (r_s) for snow and ice land use classes to 10^4 s m^{-1} . This corresponds to total deposition velocities of $\leq 0.01 \text{ cm s}^{-1}$, which is a reduction of

225 ~66% compared to the Wesely deposition routine that is the default being applied in WRF ([Grell et al., 2005](#)). Effects of this modification are further examined in Sect. 3.1.

2.3 Observational data of surface ozone

The new modelling setup, including nudging to ECMWF ERA5 and the revised O₃ deposition to snow, ice and oceans, is evaluated against observational data of pan-Arctic surface O₃ [concentrations](#). We expect that the different representation of
230 O₃ deposition mostly affects O₃ concentrations in the ABL. Therefore, we evaluate our simulations against hourly averaged surface O₃ observations from 25 measurement sites above 60 °N. These sites are further categorized in three site selections: 'High Arctic', 'Terrestrial' and 'Remote'. High Arctic refers to sites having latitudes > 70 °N and for which we expect that the deposition footprint is a combination of ocean and ~~(sea-)ice~~[sea-ice](#) (e.g. [Helmig et al., 2007b](#)). The Terrestrial sites are located below 70 °N and show a clear diurnal cycle in observed O₃ (e.g. [Chen et al., 2018](#)). These diurnal cycles are governed by
235 a combination of emissions of precursors, but also the anticipated larger diurnal cycle in O₃ deposition ([Zhou et al., 2017](#)) to, e.g., vegetated surfaces and a stronger diurnal cycle in turbulent mixing conditions and ABL dynamics. These are in all aspects different from sites that have an ocean/~~ice~~[sea-ice](#) footprint where we expect low emissions of precursors, no clear diurnal cycle in O₃ deposition and a weaker diurnal cycle in ABL dynamics ([Van Dam et al., 2015](#)). In this study, the criterion is that the average observed minimum nighttime mixing ratio is > 8 ppb smaller than the average observed maximum daytime
240 mixing ratio during the ~1 month of simulation. This criterion is based on a preparatory analysis of the observational data, footprint and site characteristics. The Remote sites ~~are located~~[have been identified as such based on their location](#) below 70 °N and ~~at which showing~~ no clear diurnal cycle ~~is observed in~~ [O₃ concentrations](#). The analysis also includes the observations during the Arctic Summer Cloud Ocean Study (ASCOS) campaign, when the icebreaker Oden was located in the Arctic sea ice (Tjernstrom et al., 2012). In total, 25 surface O₃ measurement sites are included (Fig. 1) of which 6, 8 and 11 sites are
245 characterized High Arctic, Remote and Terrestrial sites, respectively. A full list of available measurement sites is available in Tab. D1.

2.4 Overview of performed simulations

In total, we perform three simulations. The first WRF simulation (DEFAULT) is a run without any adjustments to the code as described in Sect. 2.1. The second simulation (NUDGED) includes nudging of the synoptic conditions to the ECMWF ERA5
250 product as described in Sect. 2.1.1. The third simulation (COAREG) includes nudging, but also includes the adjustments to the O₃ deposition to oceans as described in Sect. 2.2 and the O₃ deposition to snow and ice as described in Sect. 2.2.1. Furthermore, we also compare our results with the the state-of-the-art CAMS global reanalysis data product (Inness et al., 2019). This product has a temporal resolution of 3 hours, a spatial resolution of 0.75° × 0.75° ~~;~~ and does not include a mechanistic representation of ocean-atmosphere O₃ exchange. ~~Regarding O₃,~~ CAMS assimilates satellite observations [of O₃](#) but it does not assimilate
255 O₃ observations from [radiosondes or](#) in situ measurement sites ~~or radiosondes~~[such as the 25 sites used in the here presented evaluation](#). Moreover, CAMS is being widely used for air quality forecasts and assessments but also to constrain regional scale modelling experiments such as presented in this study.

3 Results

260 First, we will present the spatial and temporal variation in O_3 dry deposition velocities (V_{d,O_3}) of the ~~new and default~~ NUDGED and COAREG modelling setup including the effect on the total O_3 deposition budget. Subsequently we will discuss the resulting effect on the spatial distribution of the mean ~~background~~ surface O_3 mixing ratios. Then, we will present the comparison of all WRF simulations and CAMS data with the hourly surface observations for the three site selections (High Arctic, Remote and Terrestrial). This section is finalized by the simulated and observed time series for the six High Arctic sites.

3.1 Dry deposition budgets and distribution

265 Figure 3a and Fig. 3b show the mean deposition velocities for the ~~DEFAULT~~ NUDGED and COAREG runs, respectively. As expected, in the ~~DEFAULT~~ NUDGED run (Fig. 3a) the mean V_{d,O_3} to oceans are in the order of 0.05 cm s^{-1} . Furthermore, the spatial distribution shows a relatively low heterogeneity and no increase in deposition velocities towards the warmer waters. ~~In the~~ The COAREG run (Fig. 3b) ~~we find provides a~~ mean V_{d,O_3} in the order of 0.01 cm s^{-1} for the ~~colder waters Arctic~~ ocean $> 70^\circ\text{N}$ up to 0.018 cm s^{-1} for the warmer waters. There also appears to be an enhancement of oceans with high Γ_{aq} concentrations (Fig. C1). Simulated oceanic O_3 deposition ~~to is elevated in~~ coastal waters (e.g. Baltic Sea and around the Bering Strait) with Γ_{aq} concentrations reaching up to 130 nM compared to ~~30-30-50~~ $30-50 \text{ nM}$ for the open Arctic ocean waters (~~not shown here~~ Fig. C1). This highlights the sensitivity of the COAREG scheme to chemical enhancement with dissolved iodide.

270 Figure 3c shows the temporal variability in V_{d,O_3} for one of the grid boxes, which is in terms of temporal variability representative for the whole domain. The temporal variability in the ~~DEFAULT run is~~ NUDGED run is mainly governed by temporal variability in r_a . During episodes with high wind speeds ($> 10 \text{ m s}^{-1}$), r_a becomes so small that it is negligible over the constant surface uptake resistance of 2000 s m^{-1} , corresponding to a maximum V_{d,O_3} of 0.05 cm s^{-1} . During episodes with low wind speeds ($< 5 \text{ m s}^{-1}$), reduced turbulent transport poses some additional restriction on O_3 removal with increasing r_a which ~~can reduce~~ reduces the V_{d,O_3} ~~up to $\sim 8\%$~~ to $\sim 0.04 \text{ cm s}^{-1}$. In the COAREG run, ~~the~~ temporal variability in ~~O_3 deposition~~ V_{d,O_3} is also governed by wind speeds that controls the waterside turbulent transport of O_3 in seawater besides atmospheric turbulent transport. For high wind speeds, the waterside turbulent transport increases (Fig. B1) and more O_3 is transported through the turbulent layers. For our simulation, we found that the temporal variability in O_3 deposition due to waterside turbulent transport can be up to $\pm 20\%$ around the mean. Overall, the V_{d,O_3} to oceans in the COAREG run is reduced by $\sim 60-80\%$ compared to the ~~DEFAULT~~ NUDGED run. The mean V_{d,O_3} to snow and ice is reduced by ~~~ 30~~ 66% , from $\sim 0.03 \text{ cm s}^{-1}$ in the ~~DEFAULT~~ NUDGED run to $\sim 0.01 \text{ cm s}^{-1}$ in the COAREG run.

285 The temporal evolution in oceanic O_3 deposition velocities simulated by the COAREG run appears to be on the low side of observed and elsewhere simulated V_{d,O_3} (e.g. Chang et al., 2004; Oh et al., 2008; Ganzeveld et al., 2009). Chang et al. (2004) showed that V_{d,O_3} can increase by a factor of 5 with wind speed increasing from 0 to 20 m s^{-1} . Luhar et al. (2017) (Figure 7) shows a wide range of observed and simulated sensitivities to wind speed. Observations from the TexAQS06 summer campaign in the Gulf of Mexico show a large sensitivity to 10-meter wind speeds even though the model seems unable to
290 capture these high deposition velocities at high wind speeds (Luhar et al., 2017). However, Luhar et al. (2017) also shows

that for the GasEx08 campaign in the cold Southern Ocean the sensitivity of observed and simulated V_{d,O_3} to 10-meter wind speeds is very limited. This limited sensitivity is most accurately represented by the newer two-layer reactivity scheme compared to the older one-layer scheme due to a more limited interaction between chemical reactivity and waterside turbulent transport (Luhar et al., 2017). Furthermore, the variability around the mean presented in Tab. 1 ($0.012 \pm 0.002 \text{ cm s}^{-1}$) seems to correspond to Oh et al. (2008) ($0.016 \pm 0.0015 \text{ cm s}^{-1}$) 1 month simulation including O_3 removal by Γ_{aq} . In this study we show the intramonthly variability in oceanic O_3 deposition which is expected to be relatively low compared to the seasonal variability which will also be driven by temporal changes in solubility and reactivity due to the seasonal changes in SST and Γ_{aq} .

By estimating the total deposition flux for the water, snow/ice and land surfaces we can quantify the total simulated O_3 deposition budget (Tab. 1) for the Arctic modelling domain. Land, not covered with snow or ice, is with 48% the dominant surface type for this specific domain setup in summer. Combined with a relatively high simulated V_{d,O_3} of $\sim 0.45 \text{ cm s}^{-1}$ this is the most important sink, in terms of deposition, of simulated O_3 with $\sim 136\text{--}135 \text{ Tg } O_3 \text{ yr}^{-1}$. The simulated O_3 deposition budget to water bodies, covering 37% of the total surface area, is in the DEFAULT contributes in the NUDGED run $\sim 10\%$ ($\sim 15.5\text{--}15.4 \text{ Tg } O_3 \text{ yr}^{-1}$) of to the total O_3 deposition sink. In the COAREG run, this reduces to only $\sim 3\%$ ($\sim 4.6 \text{ Tg } O_3 \text{ yr}^{-1}$) of the total O_3 deposition sink. Simulated O_3 deposition to snow and ice, covering 15% of the total surface area, is the least important deposition sink with ~ 4.1 and $\sim 1.7 \text{ Tg } O_3 \text{ yr}^{-1}$ for in the DEFAULT and COAREG runs respectively.

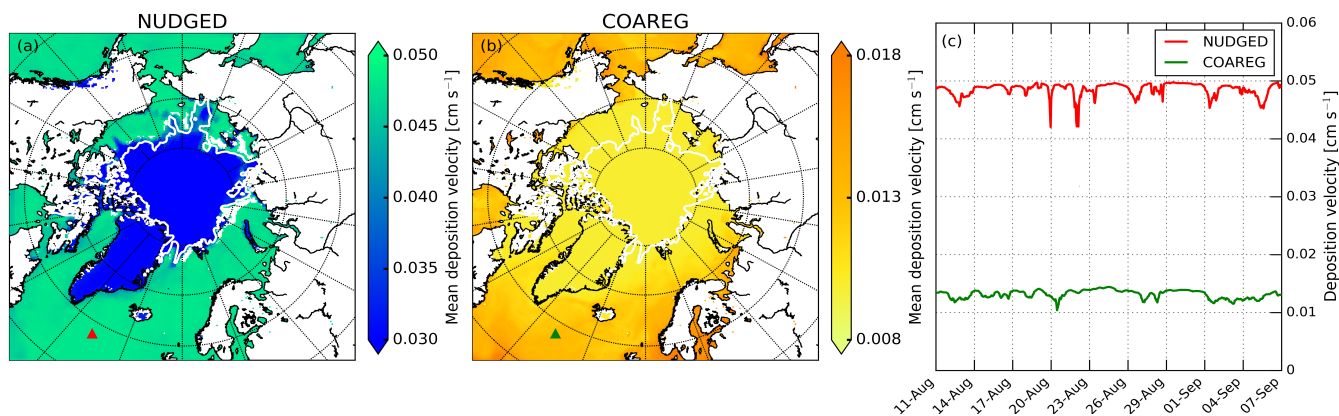


Figure 3. Spatial distribution of the mean simulated O_3 deposition velocity to snow/ice and oceans [cm s^{-1}] for the (a) ~~DEFAULT~~ NUDGED and (b) COAREG simulations and (c) temporal variation in O_3 deposition velocity [cm s^{-1}] for the ~~DEFAULT~~ NUDGED (red) and COAREG (green) simulations. The red and green markers in (a) and (b) indicate the location of the time series shown in (c). To give an indication of the sea ice extent, the white contours show the sea ice fraction of 0.5 at the start of the simulation.

3.2 Simulated and observed monthly mean surface ozone

The reduction in O_3 deposition to water and snow/ice surfaces, comparing the DEFAULT and COAREG simulation results (Sect. 3.1, Tab. 1), appears to be limited in terms of relative changes in V_{d,O_3} and the total simulated O_3 deposition budget.

Table 1. Mean simulated O₃ deposition velocity (\pm Standard deviation) [cm s⁻¹] and total simulated deposition budget [Tg O₃ yr⁻¹] for the ~~DEFAULT~~-NUDGED and COAREG runs to water, snow/ice and land each representing 37%, 15% and 48% of the total surface area respectively. The standard deviation gives an indication of the spatiotemporal variability in simulated O₃ deposition velocities.

		Water (37%)	Snow/Ice (15%)	Land (48%)	Total (100%)
NUDGED	Deposition velocity (\pm Std.) [cm s ⁻¹]	0.048 <u>0.047</u> (\pm 0.003)	0.030 (\pm 0.000)	0.449 (\pm0.23 <u>\pm0.225</u>)	
	Deposition budget [Tg O ₃ yr ⁻¹]	15.5 <u>15.4</u>	4.1	132.9 <u>133.4</u>	152.5 <u>152.9</u>
COAREG	Deposition velocity (\pm Std.) [cm s ⁻¹]	0.012 (\pm 0.002)	0.010 (\pm 0.000)	0.448 (\pm 0.251)	
	Deposition budget [Tg O ₃ yr ⁻¹]	4.6	1.7	135.8	142.1

310 ~~Especialy contrasting this with the previously mentioned up to \sim 2 orders of magnitude larger V_{d,O_3} to vegetation. However, these relatively small changes do significantly affect the spatial and temporal variation of simulated surface O₃ mixing ratios.~~ Figure 4 shows the spatial distribution in the simulated mean surface O₃ mixing ratios overlain with the observed mean surface O₃ mixing ratios. In the ~~DEFAULT~~-NUDGED and COAREG runs (Fig. 4a and Fig. 4b respectively) we find similar ~~background surface~~ background surface O₃ mixing ratios of \sim 15-20 ppb over the Russian and Canadian/Alaskan land masses. Over Scandinavia, slightly higher ~~background surface~~ background surface O₃ mixing ratios of \sim 20-25 ppb are simulated due to more anthropogenic emissions of precursors in the EDGAR emission inventory and advection of O₃ and its precursors from outside the domain. ~~We As expected, we~~ find a limited effect of reduced deposition to water and snow/ice to the simulated mean O₃ mixing ratios over land. In general, the model appears to ~~be able to~~ simulate the mean observed surface O₃ mixing ratios for the Remote and Terrestrial sites (all sites < 70 °N) generally well without clear positive or negative bias. ~~However, we~~ Due to the altitude effect higher surface O₃ concentrations ~~are simulated over Greenland even though the deposition velocity to snow and the surrounding oceans is of similar magnitude (\sim 0.01 cm s⁻¹).~~

320 The reduced O₃ deposition to water and snow/ice surfaces, comparing the NUDGED and COAREG simulation results (Sect. 3.1, Tab. 1), appears to be limited in terms of relative changes in V_{d,O_3} and the total simulated O₃ deposition budget. However, these relatively small changes do substantially affect the simulated spatial distribution of surface O₃ mixing ratios over oceans and sea ice as indicated in Fig. 4. We find that the ~~DEFAULT~~-NUDGED run (Fig. 4a) systematically underestimates the mean observed surface O₃ mixing ratios for the High Arctic sites (all sites > 70 °N) by \sim 5-10 ppb ~~likely which appears to be~~ caused by an overestimated deposition to ocean, snow and ice surfaces. ~~also further substantiated by the following analysis of short-term variability in O₃ concentrations (Sect. 3.3).~~ Over the Arctic sea ice and oceans the ABL is typically very shallow and atmospheric turbulence is relatively weak. This suppresses vertical mixing and entrainment of O₃ rich air from the free

330 troposphere. Dry deposition of O₃ to the ocean or snow/ice surfaces appears to be an important removal mechanism that has a large impact on O₃ concentrations in these shallow ABLs (Clifton et al., 2020a) both in terms of magnitude but also temporal variability as we will show in Sect. 3.4. In the COAREG run, the ~~background surface~~ background surface O₃ mixing ratios over oceans and Arctic sea ice have increased up to 50%. Furthermore, the reduced deposition to snow/ice has also clearly affected simulated surface O₃ mixing ratios over Greenland. Most importantly, the negative bias in simulated surface O₃ mixing ratios is reduced in the

335 COAREG run with respect to the ~~DEFAULT run. This is further examined in NUDGED run (see Sect. 3.3).~~ The CAMS

reanalysis data appears to simulate higher (up to 10 ppb) surface O_3 mixing ratios over land than the two WRF runs. Over sea ice, the magnitude of simulated surface O_3 mixing ratios in CAMS is in between the DEFAULT and COAREG runs. Over Greenland, CAMS simulates mixing ratios of ~ 40 ppb, with a sharp gradient towards the coast. This gradient is less pronounced in the WRF simulations. Both CAMS and WRF appear to underestimate the mean observed (~ 45 ppb) surface O_3 mixing ratios at Summit. The frequency distributions (Fig. 4d) also show that relatively high (25–45 ppb) simulated surface O_3 mixing ratios are more frequent in COAREG and CAMS compared to the DEFAULT and NUDGED runs.

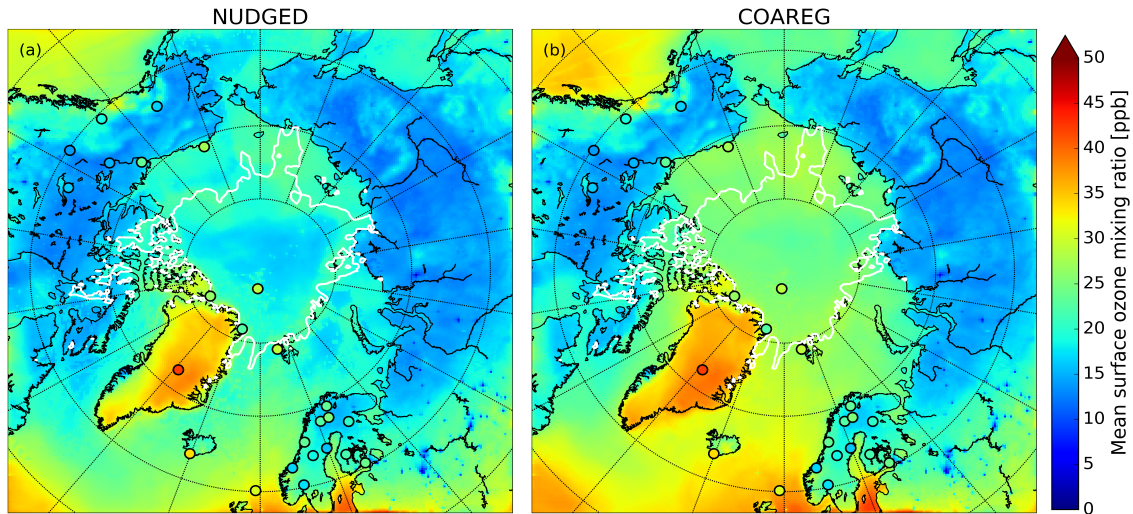


Figure 4. Spatial distribution of the simulated mean surface O_3 mixing ratio [ppb] for the (a) ~~DEFAULT~~ NUDGED and (b) COAREG runs ~~and~~ (c) CAMS data and (d) frequency distributions of surface O_3 mixing ratios ppb over the entire simulation and domain for the DEFAULT (red), NUDGED (yellow), COAREG (green) runs and CAMS data (blue). The filled circles indicate the mean observed ozone mixing ratios [ppb] for the simulated period. To indicate the sea ice extent, the white contours show the sea ice fraction of 0.5 at the start of the simulation.

3.3 Simulated and observed hourly surface ozone

In this section we show how both nudging and the application of the revised deposition scheme ~~can especially improve short-term variability in~~ improves the model prediction scores of surface O_3 concentrations reflected in a comparison of the simulated and observed hourly surface O_3 mixing ratios at the three site selections (High Arctic, Remote and Terrestrial). This is according to our knowledge the first time such ~~a~~ an oceanic O_3 deposition scheme coupled to a meteorology-chemistry model is evaluated against a large dataset of hourly surface O_3 observations. Figure 5 shows a comparison between observed and simulated hourly surface O_3 mixing ratios subdivided in the three site selections: High Arctic, Remote and Terrestrial. For As expected, for the High Arctic sites (Fig. 5, top row) we ~~again~~ find that the DEFAULT run is underestimating the observed surface O_3 mixing ratios with a mean bias of -7.7 ppb. ~~This is consistent to findings in Fig. 4, where the DEFAULT run appears~~

~~to underestimate surface O₃ mixing ratios in the High Arctic region.~~ Interestingly, nudging to ERA5 wind, temperature and humidity appears to already reduce some of the bias in the High Arctic by better representing the temporal variability in surface O₃. This is further examined in Sect. 3.4. However, the NUDGED run appears to still underestimate High Arctic surface O₃ with a bias of -3.8 ppb ~~-which is also consistent with the findings in Fig. 4, where the NUDGED run appears to underestimate~~
355 ~~surface O₃ mixing ratios in the High Arctic region.~~ The COAREG run, having a reduced O₃ deposition sink to oceans and snow/ice appears to better represent the ~~background~~-surface O₃ observations with a slight positive bias of 0.3 ppb. The MAE in the COAREG run is reduced to 4.7 ppb from 8.5 and 6.4 for the DEFAULT and NUDGED runs respectively. Furthermore, we find that the CAMS reanalysis data also underestimates surface O₃ in the High Arctic with a bias of -5.0 ppb and a MAE of 6.8 ppb. ~~It has to be noted~~ Note that the performance for all WRF runs and CAMS reanalysis product is varying for each
360 observational site which is further examined in Sect. 3.4.

For the Remote sites (Fig. 5, middle row), having no clear diurnal cycle in surface O₃, we find again an improvement by nudging the WRF model to ERA5 and also by including the mechanistic ocean deposition routine and reduced snow/ice deposition. This improvement appears to be most pronounced for coastal sites like Storhofdi (63.4°N,20.3°W) and Inuvik (68.4°N,133.7°W) ~~having with~~ a reduction in the MAE of 57% and 36% respectively (not shown here). Overall, the improve-
365 ment for the NUDGED and COAREG runs compared to the DEFAULT run in the Remote site selection is not as significant compared to the High Arctic sites, ~~probably also~~ because of the larger role of O₃ deposition to land and vegetation, which remained unchanged in this study. We find that the CAMS data shows the best performance for the Remote sites with no bias and with a MAE of 5.6 ppb.

For the Terrestrial sites (Fig. 5, bottom row), having a clear diurnal cycle in surface O₃, all WRF runs slightly overestimate
370 the observed surface O₃ mixing ratios with a mean bias up to 1.0 ppb. By nudging WRF to ERA5 the bias is reduced from 7.0 ppb to 6.0 ppb. Reducing the O₃ deposition to oceans and snow/ice increases the bias, but the MAE remains unchanged. The CAMS reanalysis data appears to perform worst for the Terrestrial sites with a bias of 6.4 ppb and a MAE of 8.0 ppb. This might be explained by the lower spatial and temporal resolution ~~in the dataset of CAMS specifically~~ at these sites ~~with~~
375 ~~having~~ a relatively strong diurnal cycle in ABL dynamics ~~and O₃ concentrations.~~ ~~Interestingly, of all the combinations, we find~~
~~the largest MAE (8.5 ppb) for the High Arctic sites in the DEFAULT run (Fig. 6a) while we find the lowest MAE (4.7 ppb)~~
~~for the High Arctic sites in the COAREG run (Fig. 6c). This indicates the high sensitivity of the adjusted ocean, snow and ice~~
~~surfaces deposition representation to the magnitude and temporal variability in surface O₃ at high latitudes. Because these sites~~
~~are located far away from the domain boundaries we expect that these model results are to a lesser extent influenced by the~~
~~boundary conditions compared to the Terrestrial and Remote sites and therefore more sensitive to the~~ O₃ deposition scheme
380 ~~in WRF to vegetation and O₃ concentrations. Also a misrepresentation of emissions of precursor emissions and concentrations~~
~~and the O₃ deposition to vegetation (Michou et al., 2005; Val Martin et al., 2014) might explain some of the differences.~~

3.4 ~~Temporal~~ Short-term temporal variability of surface ozone in the High Arctic

In Sect. 3.3 we have shown how nudging the WRF model to ERA5 synoptic conditions and revising the O₃ deposition scheme to oceans and snow/ice can improve the model's capability to represent the observed hourly surface O₃ mixing ratios, especially

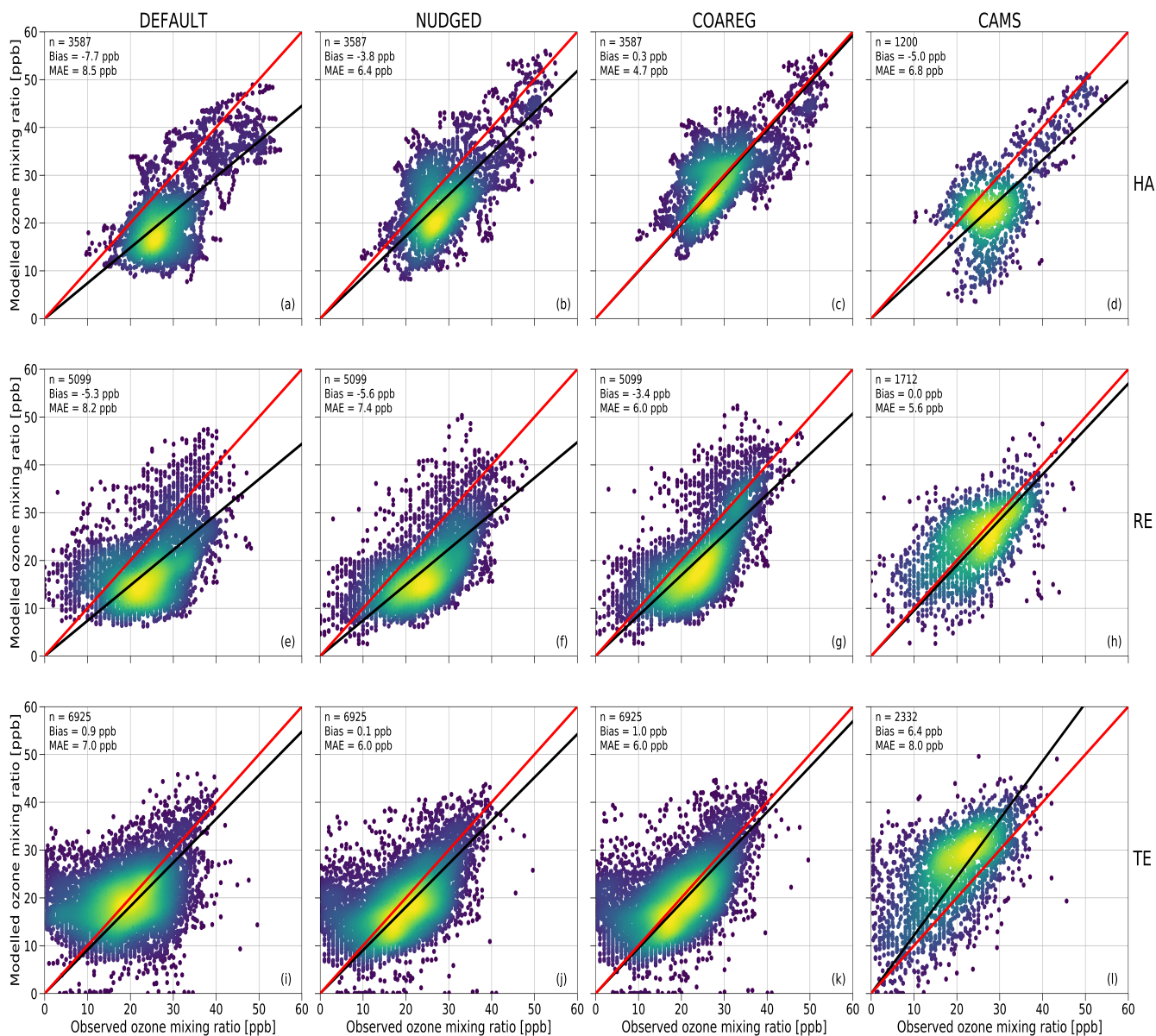


Figure 5. Comparison of the hourly observed and simulated ozone mixing ratios [ppb] for the DEFAULT (a,e,i), NUDGED (b,f,j), COAREG (c,g,k) runs and CAMS data (d,h,l) for the High Arctic (HA) (a-d), Remote (RE) (e-h) and Terrestrial (TE) (i-l) sites. The red line indicates the 1:1 line and the black line indicates the Ordinary Least Squares regression line through the origin. The number of data points (n), Bias [ppb] and Mean Absolute Error (MAE) [ppb] are shown in the top left corner. The colors represent the multivariate kernel density estimation with yellow colors having a higher density.

385 for the High Arctic sites. In this section we show how the ~~three WRF-NUDGED and COAREG~~ runs and CAMS represent the temporal variation in High Arctic surface O₃ observations, focusing on a ~~selection-6 out~~ of the 25 measurement sites. These 6 High Arctic sites have been selected due to their deposition footprint being dominated by transport over, and deposition to, ocean and sea-ice covered surfaces. Figure 6 shows the observed and simulated surface O₃ time series for ~~the 6 High Arctic (>70°N) sites:~~ ASCOS, Summit, Villum, Zeppelin, Barrow and Alert. Furthermore, Tab. 2 shows the model skill indicators for
390 the High Arctic sites. These skill indicators include the Mean Absolute Error (MAE) that represents the systematic error, the Standar Deviation of Observation minus model Prediction σ_{o-p} that represents the random error and the Pearson-R correlation coefficient (R) that represents the degree of correlation.

The observations at ASCOS (Fig. 6a) show a sudden increase of surface O₃ mixing ratios from 20 to over 30 ppb around the 17th of August due to advection of relatively ozone rich air during a synoptically active period (Tjernstrom et al., 2012).
395 Only the COAREG run appears to be able to simulate a similar increase in surface O₃ while NUDGED and CAMS show a minor increase ~~and the DEFAULT run shows no increase~~ in simulated surface O₃ ~~at all~~. From the 17th of August onwards, the observations show mixing ratios between 25 and 35 ppb. The WRF simulations indicate advection of air over ocean and ice surfaces during this time period (not shown here). In the COAREG simulation, with less deposition to these surfaces, surface O₃ mixing ratios are less depleted. Only the COAREG run is able to represent these observed mixing ratios with a bias of -2.0
400 ppb whereas the ~~other models simulate~~ NUDGED and CAMS are clearly biased towards lower mixing ratios.

At Summit (Fig. 6b), we find a large temporal variability in observed surface O₃ between 30 and 55 ppb. From the 11th of August onwards we find a decreasing trend in observed surface O₃ down to 30 ppb before increasing to 40 ppb around the 17th of August. ~~We find that the DEFAULT run is unable to~~ All models capture this specific event ~~whereas the NUDGED and COAREG runs already appear to capture this event much better~~ in terms of temporal variability even though ~~the model is~~
405 NUDGED and COAREG are still biased at the observed minimum of 30 ppb. Furthermore, we find that the CAMS reanalysis data represents this specific period very well, also in terms of magnitude. At Summit, the increase of surface O₃ in the COAREG run relative to the NUDGED run mostly reflects the reduction of deposition to snow and ice due to the prevailing katabatic wind flow (Gorter et al., 2014). During episodes with low wind speeds the ABL becomes very stable and shallow during which deposition to snow and ice becomes an important process in removing O₃ in the ABL. In the period between the 14th and 26th
410 of August this reduction in deposition can increase the surface O₃ mixing ratios up to 10 ppb (e.g. 23th of August). In contrast, during episodes with higher wind speeds and deeper ABLs the reduced O₃ deposition to snow hardly affects the simulated surface O₃ concentrations. Interestingly, we find that the NUDGED and COAREG simulations show a larger negative bias (~5-10 ppb) during the period with low wind speeds and shallow ABLs. Over the entire simulated period, CAMS performs best at Summit with a MAE of 3.9 ppb followed by COAREG with a MAE of 6.1 ppb. ~~Interestingly,~~

415 Villum (Fig. 6c) is the only site for which the ~~DEFAULT run performs best in terms of bias and MAE. This run slightly underestimates the observed mixing ratios with a bias of -2.4 ppb.~~ The NUDGED and COAREG runs as well as the CAMS reanalysis data all systematically overestimate the observed mixing ratios, especially later into the simulation. The observations show an increase in O₃ mixing ratios from 10 to 20 ppb in the first three days of the simulation where after it remains between 20 and 30 ppb with relatively low temporal variability compared to some of the other sites (e.g. Summit, Barrow). Both the

420 NUDGED and COAREG runs simulate mixing ratios up to 40 ppb and CAMS simulates maximum surface O₃ mixing ratios of 35 ppb. In terms of representing the magnitude of surface O₃ mixing ratios CAMS performs best with a MAE of 4.5.
Zeppelin (Fig. 6d) and Barrow (Fig. 6e) show similar behaviour in terms of observation-model comparison. For both locations ~~, both the DEFAULT run as well as the~~ the CAMS reanalysis data systematically ~~underestimate~~ underestimates observed ozone mixing ratios with ~~biases larger than a~~ biases > 10 ppb. In the NUDGED run ~~, some of the temporal variability is already better~~
425 ~~represented by WRF and reduces the bias to~~ the bias equals -6.9 and -4.6 ppb for Zeppelin and Barrow, respectively. In the COAREG run the bias is reduced to -1.0 and -0.2 ppb for Zeppelin and Barrow respectively. ~~From the~~ This reduction in bias is, together with ASCOS, the largest among the 6 High Arctic sites and shows the large sensitivity to the representation of O₃ deposition. At Barrow, the dominant wind directions during the simulation period are NW-NE giving a footprint mostly from the Arctic sea ice and ocean. Especially in the period from the 23th of August until the end of the simulation, we find
430 ~~a good example of the importance of a realistic representation of synoptic conditions by nudging and the role of ocean and snow/ice deposition. In this period, the DEFAULT run and CAMS reanalysis data systematically underestimate the~~ th of August onward the COAREG run is very accurate in representing the magnitude as well as the temporal variability in observed surface O₃ mixing ratios. Moreover, the COAREG run is representing the observed. During this period, the NUDGED run simulates surface O₃ mixing ratios ~~very well, both in terms of magnitude as well as temporal variability up to 5 ppb lower due~~
435 ~~to the overestimated deposition to oceans and sea ice. At both sites, the model performance of COAREG is in the same order of magnitude with an MAE, σ_{o-p} and R of \sim 3.5 ppb, 4.2 ppb and 0.65 respectively.~~
At Alert (Fig. 6f), ~~the DEFAULT run again underestimates the~~ we find a relatively steady increase in observed surface O₃ ~~mixing ratios even though the bias of -6.4 ppb is not as large as for~~ from 20 ppb at the start of the simulation to 30 ppb at the end of the simulation. The temporal variability, both in observed and simulated surface O₃ appears to be lower compared to
440 ~~some of the other sites~~ High Arctic sites. Again, the statistical parameters such as MAE, σ_{o-p} and R improve in the COAREG run with respect to the NUDGED run. This bias, as well as the MAE, is again decreased for the NUDGED and COAREG runs.
At Alert, we find that CAMS has the lowest MAE and σ_{o-p} of 3.0 ppb ~~, but has a slight negative bias of -1.9 ppb and 3.4 ppb~~ respectively.
The model performance in terms of temporal variability in surface O₃ observations is diagnosed by using the Pearson-R
445 correlation coefficient. ~~Nudging the WRF model to ERA5 meteorological data already improved the representation of the temporal variability especially for sites like Barrow and Summit where the synoptic conditions were likely not represented well. This causes an offset in timing of the advection of different air masses but here also vertical mixing and entrainment of O₃ rich air could play a role.~~ The model performance ~~also~~ improved for all six sites in the COAREG run with respect to the NUDGED run. The COAREG run includes temporal variability in O₃ deposition due to variability in waterside turbulent transport which
450 can explain additional improvements in representing the temporal variability of surface O₃. The COAREG simulation performs best for 5 out of the 6 observational sites in terms of Pearson-R correlation coefficient and is only outperformed by CAMS at Summit. Overall, we find that ~~nudging reduces the bias and MAE for all High Arctic sites except Summit and Villum by better representing the synoptic conditions and therefore the temporal variability in observed surface O₃. Coupling the~~ coupling the WRF model to the mechanistic COAREG ocean-atmosphere exchange representation ~~further decreases the bias and MAE~~

455 decreases the MAE and σ_{O_3} for all High Arctic sites except for Villum by better representing the magnitude of, but also temporal variability in observed surface O₃. The CAMS reanalysis data is performing well for some locations (e.g. Summit, Alert) while for Zeppelin and Barrow the discrepancy is among the largest we found in the observation-model comparison.

Table 2. Bias-MAE [ppb], MAE- σ_{O_3} [ppb] and Pearson-R correlation coefficient (R) [-] for the ~~DEFAULT~~, NUDGED, and COAREG runs and CAMS reanalysis data at the ASCOS, Summit, Villum, Zeppelin, Barrow and Alert observational sites. The lowest model error and highest correlation have been made bold for every site.

	ASCOS				Summit		
	Bias-MAE	σ_{O_3}	R	Bias-MAE	σ_{O_3}		
DEFAULT -11.5 11.5 0.24 -5.3 7.4 0.17 - 2.4 4.5 0.5 -9.5 9.5 0.61 -12.4 12.4 -0.18 -6.4 6.6 0.43-NUDGED	-9.4	9.4	<u>4.3</u>	0.46	-5.5	7.5	<u>7.0</u>
COAREG	-2.0	3.1	3.2	0.67	-4.0	6.1	<u>5.8</u>
CAMS	-6.8	7.5	<u>4.5</u>	0.07	-2.6	3.9	<u>4.3</u>

4 Discussion

460 ~~In this study, we demonstrate the role~~This study demonstrates the impact of a mechanistic representation of ocean-atmosphere O₃ exchange to simulate the magnitude and temporal variability of hourly surface O₃ concentrations in the Arctic region. We show that the ~~model modelled~~ sensitivity of the surface O₃ concentrations to the representation of O₃ to ocean, ice and snow surfaces is high, even though the total deposition budget is an order of magnitude smaller than the deposition to land and vegetation. Using a mechanistic ~~representation of oceanic~~ O₃ deposition ~~to oceans and reducing the representation and reduced~~ O₃ deposition to snow and ice greatly reduced the negative bias in surface O₃, especially in the high Arctic. Furthermore, the short-term temporal variability in surface O₃ was also better represented by the mechanistic representation of oceanic O₃ deposition by also accounting for temporal variations in the driving processes of oceanic O₃ deposition such as waterside turbulent transport.

470 Our main objective was to address the ~~role impact~~ of a mechanistic oceanic O₃ deposition representation, including spatial and temporal variability, on the magnitude and temporal variability of surface O₃ concentrations and to evaluate this with a large dataset of 25 observational sites in and around the Arctic. We show that Arctic surface O₃ concentrations are ~~very~~ sensitive to the representation of O₃ deposition ~~-We did not address include in the presented analysis how the nudging and representation of Arctic O₃ deposition further affects the contribution to the Arctic O₃ budget e.g. by changes in photochemistry and stratosphere-troposphere exchange and advection. For such a budget analysis it would be best to perform at least one year of simulation to also address the seasonal cycles in deposition, photochemistry and long range transport which is computationally too expensive in WRF. Regarding oceanic~~ to oceans and sea-ice especially at coastal sites and sites with latitudes >70°N. At sites with a more terrestrial footprint (e.g. Norway, Sweden, Finland), the comparison of modelled and observed surface O₃ concentrations also shows a discrepancy. As expected, this discrepancy has not been resolved introducing the more mechanistic representation of O₃ deposition oceans and modified snow/sea-ice deposition rate. In terms of deposition, these sites are mostly

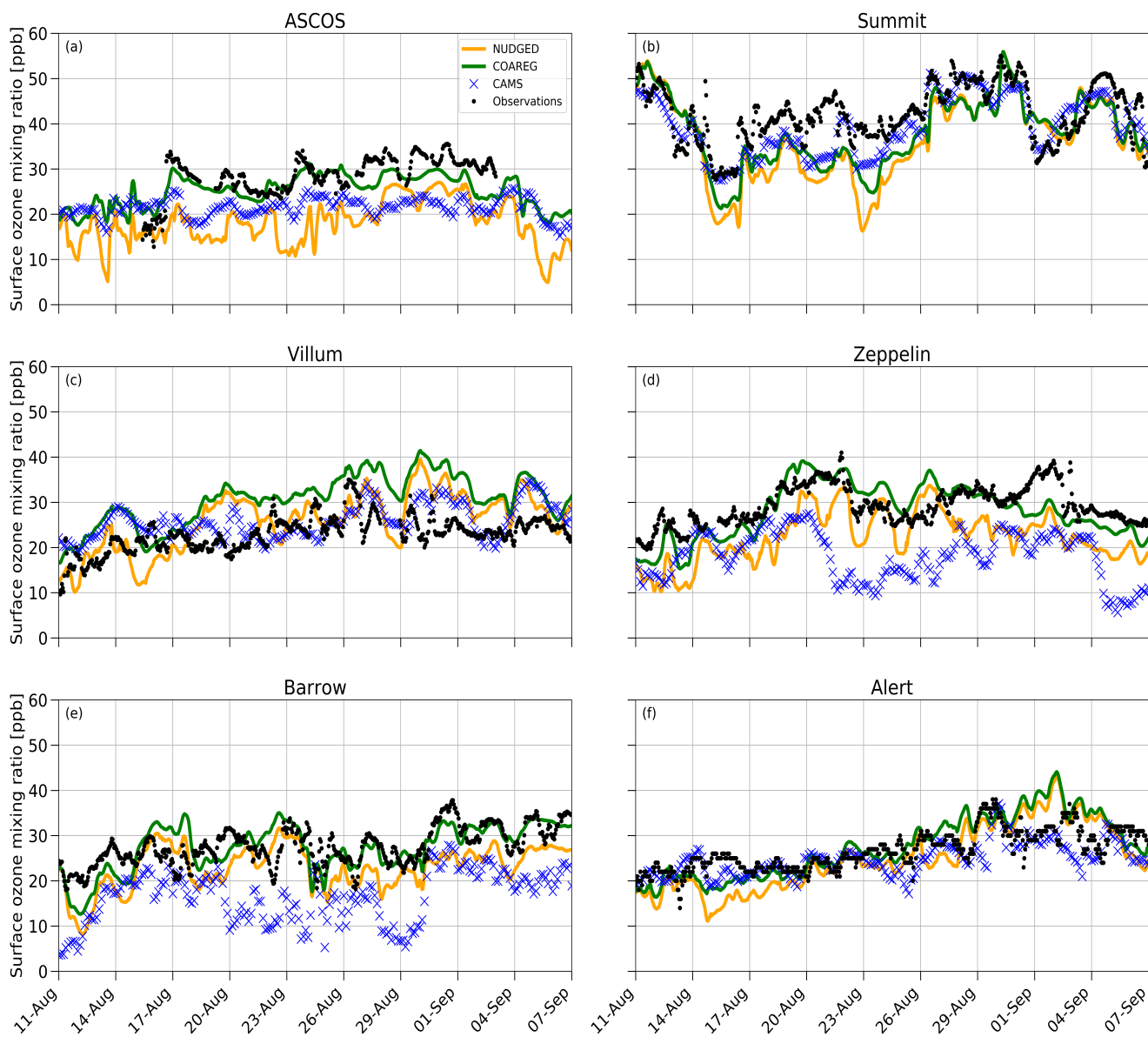


Figure 6. Temporal evolution of hourly surface O₃ mixing ratios [ppb] for the ~~DEFAULT (red)~~, NUDGED (yellow), and COAREG (green) runs, CAMS data (blue crosses) and observations (black dots) at ASCOS (~87.4°N,~6.0°W), Summit (72.6°N,38.5°W), Villum (81.6°N,16.7°W), Zeppelin (78.9°N,11.9°E), Barrow (71.3°N,156.6°W) and Alert (82.5°N,62.3°W).

influenced by O_3 deposition to vegetation and land (e.g. Silva and Heald, 2018; Wong et al., 2019; Clifton et al., 2020b). In the
480 WRF simulations, dry deposition of O_3 to vegetation (mostly land-use class: 'wooded tundra') amounts to ~ 0.2 - 1.0 cm s^{-1}
with a clear diurnal cycle. Dry deposition of O_3 to 'bare tundra' is in the order of 0.1 - 0.15 cm s^{-1} which is slightly higher
than observed by Van Dam et al. (2015). A detailed analysis of O_3 deposition ~~this would also include long-term changes
in sea ice cover and oceanic biogeochemistry. The major constraint in this model setup is the lack of oceanic to land and
vegetation is beyond the scope of this study and would require a different strategy, e.g. direct comparison with O_3 flux
485 measurements (e.g. Van Dam et al., 2016). However, a better understanding and model representation of the drivers of O_3 de-
position ~~measurements over the Arctic ocean. The COAREG exchange routine has been built and validated using~~ to vegetation
and land, including the diurnal and seasonal variability of these drivers (Lin et al., 2019), is anticipated to also result in a better
representation of short-term variability of surface O_3 over land.~~

The COAREG scheme has been developed and validated against eddy-covariance measurements over mostly (sub-)tropical
490 waters (Bariteau et al., 2010; Helmig et al., 2012). The COAREG routine has been ~~further developed and used~~ applied to
study the effects of wind speed and sea state on ocean atmosphere gas transfer (Blomquist et al., 2017; Bell et al., 2017;
Porter et al., 2020). We do expect that these main drivers, being waterside turbulent transfer and chemical enhancement with
dissolved iodide, ~~held for oceans also controls oceanic O_3 deposition~~ at high latitudes. ~~Using indirect information to evaluate~~
Indirect evaluation of oceanic O_3 deposition through comparison of surface O_3 observations instead of direct oceanic O_3 flux
495 measurements ~~we show that the addition of indicates that including~~ this mechanistic representation of O_3 deposition ~~results
in a better representation of both the improves both the modelled~~ magnitude and temporal variability in surface O_3 observa-
tions. However, ~~the exact magnitude and variability in Arctic a lack of~~ oceanic O_3 deposition ~~could not be evaluated using
flux measurements flux measurements hampers the direct model evaluation of the high-latitude O_3 deposition flux. This is
expected to be soon resolved by getting access to O_3 flux observations collected in the Multidisciplinary drifting Observatory
500 for the Study of Arctic Climate (MOSAIC) 1-year field campaign.~~

Furthermore, we have reduced the deposition to snow and ice ~~based on a study by Helmig et al. (2007a). The results of that
study also further following Helmig et al. (2007a) and (Clifton et al., 2020b). Results of Helmig et al. (2007a) also~~ motivated
follow-up observational and modelling studies aiming at the development of ~~similar to COAREG for oceanic O_3 deposition,~~
more mechanistic representations of O_3 deposition to snow/ice covered surfaces. For example, efforts have been made to
505 simulate O_3 dynamics in and above the snowpack using a 1D model setup to ~~evaluate explain~~ observations of O_3 and NO_x
concentrations measured above and inside the Summit snowpack (Van Dam et al., 2015). This 1D modelling study showed the
main role of aqueous-phase oxidation of O_3 with formic acid in the snowpack (Murray et al., 2015). Comparable 1D modelling
studies focused on assessing the role of catalytic ozone loss via bromine radical chemistry in the snowpack interstitial air
(Thomas et al., 2011; Toyota et al., 2014). However, these studies mainly ~~arrived at conclusions regarding addressed~~ the role
510 of some of this snowpack chemistry in explaining, partly observed, O_3 concentrations and not so much on snow-atmosphere
 O_3 fluxes and derived deposition rates that would corroborate the inferred very small O_3 deposition rates by Helmig et al.
(2007a). ~~Clifton et al. (2020b) summarized that accurate process-based modelling of O_3 deposition to snow requires better
understanding of the underlying processes and dependencies.~~ An eddy-covariance system has been set up as part of the MO-

SAiC campaign and will provide year-round O₃ deposition fluxes to several land surface types such as open ocean and sea ice
515 with fluctuating snow cover. These measurements will further enhance our understanding of O₃ deposition in shallow ABLs at
high latitudes ([Clifton et al., 2020a](#)) and the further role in regional atmospheric chemistry.

In this study we used the COAREG transfer algorithm version 3.6 which is extended with a two-layer scheme for surface
resistance compared to the previous versions (Fairall et al., 2007, 2011) and similar to ~~the work by Luhar et al. (2018). Oceanic
iodide (I_{aq}) is generally deemed to be the most significant reactant for O₃ in ocean water (Chang et al., 2004). Similar to
520 Pound et al. (2019) we have used the global I_{aq} distribution by Sherwen et al. (2019) on a spatial resolution of 0.125°×0.125°.
This distribution replaces the previously applied iodide estimations only using SST (Chance et al., 2014; MacDonald et al., 2014)
-Using the Sherwen et al. (2019) distribution for August/September we found relatively high I_{aq} concentrations ranging from
30 to 130 nM whereas the MacDonald et al. (2014) estimation would imply I_{aq} concentrations ranging from 5 to 50 nM. This
implies that in the WRF setup, using the Sherwen et al. (2019) I_{aq} distribution, the cold Arctic ocean is still quite effective
525 in removing O₃ from the surface waters having I_{aq} as a reactant. On the global scale, the most recent I_{aq} climatology by
Sherwen et al. (2019) most accurately represents the observed I_{aq} compared to estimations only using SST (Chance et al., 2014; MacDonal
-However, Sherwen et al. (2019) noted that the I_{aq} estimations at high latitudes (north of ≥65°N) are very poorly constrained
by the observational datasets and are therefore also an uncertainty in this study on Arctic O₃. Therefore, new I_{aq} measurements
at high latitudes, for example those performed during the MOSAiC expedition, will be very useful to better constrain the
530 global I_{aq} distributions as well as mechanistic oceanic O₃ deposition representations. The WRF simulations in this study did
not consider [Luhar et al. \(2018\)](#). Our WRF simulations excluded the additional role of chlorophyll, Dissolved Organic Matter
(DOM) or other species such as DMS on chemical enhancement of O₃ in surface waters. Experimental studies have shown that
DMS, chlorophyll, or other reactive organics, ~~can may~~ enhance the removal of O₃ at the sea surface ([Chang et al., 2004](#); [Clifford et al., 2008](#)
[Chang et al., 2004](#); [Clifford et al., 2008](#); [Reeser et al., 2009](#); [Martino et al., 2012](#)). The global modelling study by Ganzeveld
535 et al. (2009) included a chlorophyll-O₃ reactivity that increased linearly with chlorophyll concentration as a proxy for the
role of DOM in oceanic O₃ deposition. ~~The addition of this reaction significantly~~ [Including this reaction substantially](#)
enhances O₃ deposition to coastal waters such that actually observed O₃ deposition to these coastal waters is well reproduced
(Ganzeveld et al., 2009). Other studies ~~on oceanic O₃ deposition~~ such as Luhar et al. (2017); Pound et al. (2019) ~~did not
consider ignored~~ the potential role of DOM-O₃ chemistry in oceanic O₃ deposition. ~~The study by~~ Luhar et al. (2018), which
540 did not explicitly consider coastal waters, even suggested that including such a reaction deteriorates the comparison with O₃
flux observations above open oceans. A considerable uncertainty in the DOM-O₃ reaction is the second-order rate coefficient
but also the magnitude and variability in oceanic DOM concentrations (Luhar et al., 2018). To test the sensitivity of our model
setup to other reactants in the surface water we have performed an additional sensitivity analysis including the chlorophyll-O₃
and DMS-O₃ reactions from Ganzeveld et al. (2009). ~~Regarding chlorophyll we have used the monthly Oceanic chlorophyll
545 concentrations have been retrieved from the~~ 9×9 km resolution MODIS chlorophyll- α ~~e~~ concentrations dataset available at
https://modis.gsfc.nasa.gov/data/dataproduct/chlor_a.php (last access: 14 Aug 2020). ~~For DMS~~ [Chlorophyll- \$\alpha\$ concentrations are
typically < 3 mg m⁻³ for open oceans up to 25 mg m⁻³ for coastal waters. For oceanic DMS concentrations](#), we use the monthly
climatology from Lana et al. (2011). The sensitivity study with chlorophyll as extra reactant indicated a slight increase (up~~

to 5%) in deposition to coastal waters with chlorophyll concentrations up to 25 mg m^{-3} . However, the resulting effect on surface O_3 concentrations was not significant due to the large fraction of oceans with very low ($< 3 \text{ mg m}^{-3}$) chlorophyll- α concentrations. Also the reactions with oceanic DMS appear to be weak due to relatively low DMS concentrations in August/September. ~~A-These sensitivity studies indicate that I_{aq} is the main driver of chemical reactivity of O_3 in the Arctic ocean in summer. However a~~ potential sensitivity of these reactants on Arctic O_3 deposition could especially be expected in the spring to summer transition following from algal blooms (Stefels et al., 2007; Riedel et al., 2008). However, in springtime the removal of Arctic O_3 near the surface is also largely affected by halogen chemistry (Pratt et al., 2013; Thomas et al., 2013). ~~In this season, the~~ (Pratt et al., 2013; Thomas et al., 2013; Yang et al., 2020) and which is known to explain observed surface O_3 mixing ratios can drop dropping to 0 ppb (Halfacre et al., 2014). ~~It this study we have limited our analysis to a period in which~~ However, this feature is of less relevance for the presented study with the evaluation being focused on August/September and when the role of halogen chemistry is ~~not important deemed being less important~~ (Yang et al., 2020).

We nudged the WRF model to the ECMWF ERA5 reanalysis product to ensure a fair model evaluation with observations due to a better representation of the synoptic conditions. This indicated the important role of the model representation of ~~meteorolog~~meteorology, e.g. advection of polluted air and mixing/entrainment of O_3 in the ABL, in representing the observed surface O_3 concentrations. ~~An improvement in simulated synoptic conditions was also found when initializing and nudging the model with ECMWF ERA-Interim data (Dee et al., 2011). This indicates that both reanalysis products have a better representation of the actual synoptic conditions than the free running WRF model.~~ The model evaluation was set up at a resolution of $30 \times 30 \text{ km}$ which is in the order of the ERA5 reanalysis data ($0.25^\circ \times 0.25^\circ$) used for initial conditions, boundary conditions and nudging. ~~Nudging, but then to the NCEP-FNL reanalysis data, was also applied in a study by Marelle et al. (2017) using WRF for quasi-hemispheric simulations of aerosols and O_3 in the Arctic at a resolution of $100 \times 100 \text{ km}$. In this study~~ Here, we opted for a ~~$30 \times 30 \text{ km}$ setup~~ km grid spacing because we expect that the main drivers of tropospheric O_3 (chemical production and destruction, stratosphere-troposphere transport, dry deposition and mixing/advection processes) can be sufficiently resolved at this ~~resolution~~ grid spacing especially over the relatively homogeneous ocean, ice and snow surfaces. However, we do realize that ~~the use of a $30 \times 30 \text{ km}$ might have caused some issues in such a coarse grid spacing may have hampered~~ representing local air flow phenomena such as katabatic winds (Klein et al., 2001) which could explain some of the mismatch at sites like Villum (Nguyen et al., 2016). Another justification for the ~~$30 \times 30 \text{ km}$ resolution~~ km grid spacing was to limit computational time and to have a large enough domain to cover the entire region above 60°N to conduct a large pan-Arctic evaluation while at the same time having all observational sites far enough from the domain boundaries to limit the effect of the imposed meteorological and chemical boundary conditions.

~~We plan to use a similar model setup, but then at a higher resolution or using a 1D-setup, to evaluate the~~ In general, the relatively scarce Arctic observations introduces constraints to modelling studies and limits the potential of these results to be extrapolated to other seasons and lower latitudes. In this case, this includes the uncertainty in the magnitude and distribution of driving factors of oceanic O_3 concentration and flux measurements in and around the Arctic sea ice deposition such as I_{aq} (Sherwen et al., 2019) or DOM. New I_{aq} measurements at high latitudes, for example those performed during the year-round MOSAiC expedition. These observations will likely give insight in the role, will be very useful to better constrain the global

Γ_{aq} distributions as well as mechanistic oceanic O_3 deposition representations. Measurements of O_3 deposition to sea-ice and concentrations and deposition fluxes to the Arctic ocean during different seasons (e.g. wintertime with no photo-chemistry or springtime with active halogen chemistry) and for a wide range of meteorological conditions. Furthermore, this local flux and concentration evaluation can be extended to species such as DMS which is now also included in the COAREG version that is coupled to WRF. However, this lacks a combined seawater and atmospheric concentration and flux dataset to conduct a local validation or a similar pan-Arctic distributed surface network such as presented here for O_3 to perform an indirect regional assessment. can assist to better constrain these modelling setups in terms of magnitude and temporal variability and potentially indicate of the sensitivity to other environmental factors such as wind speed in waters with low reactivity. Furthermore, including the role of halogen chemistry (Pratt et al., 2013; Thompson et al., 2017) might give an indication of the combined role of halogens and oceanic deposition in removing O_3 and explaining the magnitude and short- but also long-term variability of O_3 concentrations in the High Arctic.

595 5 Conclusions

The mesoscale meteorology-chemistry model Polar-WRF-Chem was coupled to the Coupled Ocean-Atmosphere Response Experiment Gas transfer algorithm (COAREG) to allow for a mechanistic representation of ocean-atmosphere exchange of trace gases. Regarding the deposition of ozone (O_3) to ocean waters, this mechanistic representation includes the O_3 . This scheme represents effects of molecular diffusion, solubility, waterside turbulent transfer and chemical enhancement of O_3 uptake through its reactions with dissolved iodide. The new mechanistic representation GOAREG scheme replaces the constant surface uptake resistance approach often applied in ACTMs. Furthermore, we have increased the modelled O_3 surface uptake resistance to snow and ice. In total, three simulations were performed: 1) default WRF setup (DEFAULT), 2) nudged to ERA5 synoptic conditions (NUDGED) and 3) with adjustments to O_3 surface uptake resistance as described above (COAREG). Furthermore, the CAMS global reanalysis data product has also been included in the comparison to illustrate some limitations in the Arctic presented evaluation on High Arctic surface O_3 . This CAMS product is widely used in air quality assessments and to constrain regional scale modelling experiments. This provides additional information on the quality of the CAMS data products but also on potential issues in the representation of O_3 sources and sinks, e.g., oceanic and snow/sea-ice deposition, for the High Arctic. The modelling approach was set up for an one month at the end-of-summer period in 2008 and evaluated against hourly surface O_3 at 25 sites for latitudes $> 60^\circ N$ including observations over the Arctic sea ice as part of the ASCOS campaign.

Using the mechanistic representation of ocean-atmosphere exchange, O_3 deposition velocities were simulated in the order of 0.01 cm s^{-1} compared to $\sim 0.05 \text{ cm s}^{-1}$ in the constant surface uptake resistance approach. In the COAREG run, the spatial variability (0.01 to 0.018 cm s^{-1}) in the mean O_3 deposition velocities expressed the sensitivity to chemical enhancement with dissolved iodide. The temporal variability of O_3 deposition velocities (up to $\pm 20\%$ around the mean) is governed by surface wind speeds and expressed differences in waterside turbulent transport. In the constant surface uptake resistance approach, there is no spatial variability in O_3 deposition velocities and the temporal variability is determined by the aerodynamic resis-

tance term that can be significant at low wind speeds. Using the mechanistic representation of ocean-atmosphere exchange reduced the total simulated O₃ deposition budget to water bodies by ~~~70% a factor of 3.3 compared to the default constant ocean uptake rate approach~~ and the increase in surface uptake resistance to snow and ice reduced the deposition budget by ~~~60% a factor of 2.4.~~

Despite the fact that O₃ deposition to oceans, snow and ice surfaces only constitutes a small term in the total O₃ deposition budget (~~more than~~ > 90% of the deposition is to land), we find a substantial sensitivity to the simulated surface O₃ mixing ratios. In the COAREG run, the simulated mean monthly surface O₃ mixing ratios have increased up to 50% in the typically shallow Arctic ABL above the oceans and ~~(sea-)ice-sea-ice~~ relative to the ~~DEFAULT-NUDGED~~ run. The mechanistic representation of O₃ deposition to oceans, but also nudging to ERA5 synoptic conditions, resulted in a substantial improved representation of surface O₃ observations, especially for the High Arctic sites having latitudes > 70 °N. The DEFAULT run ~~was underestimating underestimated~~ the observed surface O₃ mixing ratios with a bias of -7.7 ppb whereas the NUDGED and COAREG runs had a bias of -3.8 ppb and 0.3 ppb, respectively. The evaluation of the WRF runs at individual High Arctic sites showed that using the mechanistic representation of O₃ deposition to oceans and nudging the model to ERA5 better represents the surface O₃ observations in terms of magnitude as well as short-term temporal variability. ~~The evaluation of the CAMS reanalysis product also indicated limitations to represent the observed surface O₃ at the High Arctic in terms of magnitude and temporal variability. Similar to DEFAULT and NUDGED. Similar to the NUDGED run,~~ CAMS underestimated High Arctic observed surface O₃ with a bias of -5.0 ppb indicating that for this product the deposition removal mechanism to oceans and snow/ice might also be overestimated ~~and should be reconsidered.~~

This study highlights the ~~role-impact~~ of a mechanistic representation of oceanic O₃ deposition on Arctic surface O₃ concentrations at a high (hourly) temporal resolution. It ~~mostly~~ corroborates the findings of global scale studies (~~e.g. Ganzeveld et al., 2009; Luhar et al.~~) and recommends that the representation of O₃ deposition to oceans and snow/ice in global and regional scale ACTMs should be revised. This revision is needed not only to better quantify the O₃ budget at the global scale, but also to better represent the observed magnitude and short-term temporal variability of surface O₃ at the regional scale. ~~In addition, explicit consideration of the mechanisms involved in O₃ removal by the oceans (and sea-ice/snow pack) are essential to also evaluate the role of potentially important feedback mechanisms and future trends in- and the role of O₃ in Arctic climate change as a function of declining sea ice cover, increasing emissions and changes in oceanic biogeochemical conditions.~~ On the regional scale, this study also has implications ~~on the fate of the Arctic O₃ budget for methods to quantify future trends in Arctic tropospheric O₃,~~ Arctic air pollution and climate in a period of declining sea ice and increasing local emissions of precursors. ~~Furthermore, this study also serves as a preparatory study for an extensive evaluation of the upcoming year-round Arctic O₃, and other climate active trace gases, concentration and deposition flux measurements as part of the MOSAiC campaign.~~

Code availability. The COAREG algorithm is available at ftp://ftp1.esrl.noaa.gov/BLO/Air-Sea/bulkalg/cor3_6/gasflux36/, last access: 10 September 2020. The coupled Polar-WRF-Chem model, model output and post-processing scripts are available upon request.

Author contributions. JGMB, LNG and GJS designed the experiment. JGMB performed the Polar-WRF-Chem simulations and performed
650 the analysis. JGMB, LNG, GJS and MCK wrote the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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Appendix A: WRF physical and chemical parameterization schemes.

Table A1. WRF physical and chemical parameterization schemes.

WRF option	Configuration
Physical parameterizations	
Microphysics	WSM5 (Hong et al., 2004)
Long wave radiation	RRTMG (Iacono et al., 2008)
Short wave radiation	RRTMG (Iacono et al., 2008)
Surface layer	Monin-Obukhov (Janjić, 2001)
Land surface	Noah (Chen and Dudhia, 2001)
Boundary layer	MYJ (Janjić, 1994)
Cumulus	Kain-Fritsch (Kain, 2004)
Chemistry	
Gas-phase	CBM-Z (Gery et al., 1989; Zaveri and Peters, 1999)
Photolysis	Fast-J (Wild et al., 2000)
Emissions	
Anthropogenic	EDGAR (Janssens-Maenhout et al., 2017)
Biogenic	MEGAN (Guenther et al., 2012)
Boundary conditions	
<u>Meteorology</u>	<u>ERA5 (0.25° × 0.25°) (Hersbach et al., 2020)</u>
<u>Chemistry</u>	<u>CAMS (0.75° × 0.75°) (Inness et al., 2019)</u>

Appendix B: Formulation of the air- and waterside resistance terms

660 The exchange velocity, in this case deposition, of ozone (V_{d,O_3}) [$m\ s^{-1}$] is calculated from the waterside resistance (r_w) [$s\ m^{-1}$] and air side resistance terms ($r_a + r_b$) [$s\ m^{-1}$] as follows:

$$V_{d,O_3} = \frac{1}{\alpha r_w + r_a + r_b}. \quad (B1)$$

Here, α [-] is the dimensionless solubility of O_3 in sea water calculated from SST [K] following Morris (1988) as

$$\alpha = 10^{-0.25 - 0.013(SST - 273.16)} \quad (B2)$$

and the waterside resistance term (r_w) is calculated as

$$665 \quad r_w = \sqrt{a * D} \frac{\Psi K_1(\xi_\delta) \cosh \lambda + K_0(\xi_\delta) \sinh \lambda}{\Psi K_1(\xi_\delta) \sinh \lambda + K_0(\xi_\delta) \cosh \lambda}. \quad (B3)$$

Here, a [s^{-1}] is the chemical reactivity of O_3 with Γ_{aq} calculated with the second order rate coefficient [$M^{-1}\ s^{-1}$] from Magi et al. (1997) and the Γ_{aq} concentrations [M] from Sherwen et al. (2019):

$$a = k \cdot [\Gamma_{aq}] = \exp\left(\frac{-8772.2}{SST} + 51.5\right) \cdot [\Gamma_{aq}]. \quad (B4)$$

670 In Eq. B3, D [$m^2\ s^{-1}$] is the molecular diffusivity of O_3 in ocean water and is calculated from the kinematic viscosity ν [$m^2\ s^{-1}$] and the waterside Schmidt number (S_{cw}) [-] as

$$D = \frac{\nu}{S_{cw}} = \frac{\mu}{\rho} / [\sqrt{44/48} \cdot \exp(-0.055 \cdot SST + 22.63)] \quad (B5)$$

where μ [$kg\ m^{-1}\ s^{-1}$] is the dynamic viscosity of seawater and ρ [$kg\ m^{-3}$] is the density of seawater.

Finally, the air side resistance terms ($r_a + r_b$) [$s\ m^{-1}$] of the deposition velocity in Eq. B1 are calculated as

$$r_a + r_b = [C_d^{-1/2} + 13.3 S_c^{1/2} - 5 + \frac{\log(S_c)}{2\kappa}] / u_{*,a} \quad (B6)$$

675 where C_d [-] is the momentum drag coefficient, S_{ca} [-] is the Schmidt number for ozone in the atmosphere, κ is the Von Karman constant (0.4) and $u_{*,a}$ [$m\ s^{-1}$] is the friction velocity in the atmosphere. The $r_a + r_b$ term is typically in the order of $100\ s\ m^{-1}$ (Fairall et al., 2011).

680 Compared to COAREG version 3.1 (Fairall et al., 2007, 2011), COAREGv3.6 is extended with a two-layer scheme based on Luhar et al. (2018). This extension is included in the second term of the waterside resistance term (Eq. B3). Here, $\Psi = \sqrt{1 + (\kappa u_{*,w} \delta_m / D)}$, $\xi_\delta = \sqrt{2ab(\delta_m + bD/2)}$, and $\lambda = \delta_m \sqrt{a/D}$ with $b = 2/(\kappa u_{*,w})$. This part of the equation is a function of the chemical reactivity a [s^{-1}] (Eq. B4), the waterside friction velocity $u_{*,w}$ [$m\ s^{-1}$], the molecular diffusivity of O_3 in ocean water (Eq. B5) and δ_m [m] representing the depth of the interface between the top water layer and the underlying turbulent layer. In this study we have applied $\delta_m = c_0 \sqrt{D/a}$ with $c_0 = 0.4$ based on Luhar et al. (2018). $K_0(\xi_\delta)$ and $K_1(\xi_\delta)$ are the modified Bessel

functions of the second kind of order 0 and 1, respectively. For more information on the derivation of the formulas please visit
685 [Fairall et al. \(2007, 2011\)](#); [Luhar et al. \(2018\)](#).

Figure B1 shows the sensitivity of the COAREG routine coupled to WRF to the environmental factors wind speed, SST and Iodide concentration. The sensitivity to wind speeds (Fig. B1a) expresses the role of waterside turbulent transport and aerodynamic resistance. For low wind speeds waterside turbulent transport is limited and therefore limits the exchange of O_3 from the atmosphere to the ocean. At high wind speeds, the dry deposition of O_3 is limited by chemical reactivity of O_3 with Γ_{aq} at typical Arctic SSTs of 5°C and Γ_{aq} concentrations of 60 nM (see also Fig. C1). At very low wind speeds ($< 3\text{ m s}^{-1}$) the aerodynamic resistance poses an extra restriction on the ocean-atmosphere exchange of O_3 . The sensitivity to SST (Fig. B1b) mostly represents the role of solubility (Eq. B2) with warmer waters having a lower solubility. In contrast to [Luhar et al. \(2018\)](#), the SST is not used to calculate the Γ_{aq} concentrations and does therefore not show a positive correlation. The sensitivity to Γ_{aq} (Fig. B1c) represents the role of chemical enhancement which is stronger than the generally compensating effect of solubility in warmer waters for typical Arctic conditions.

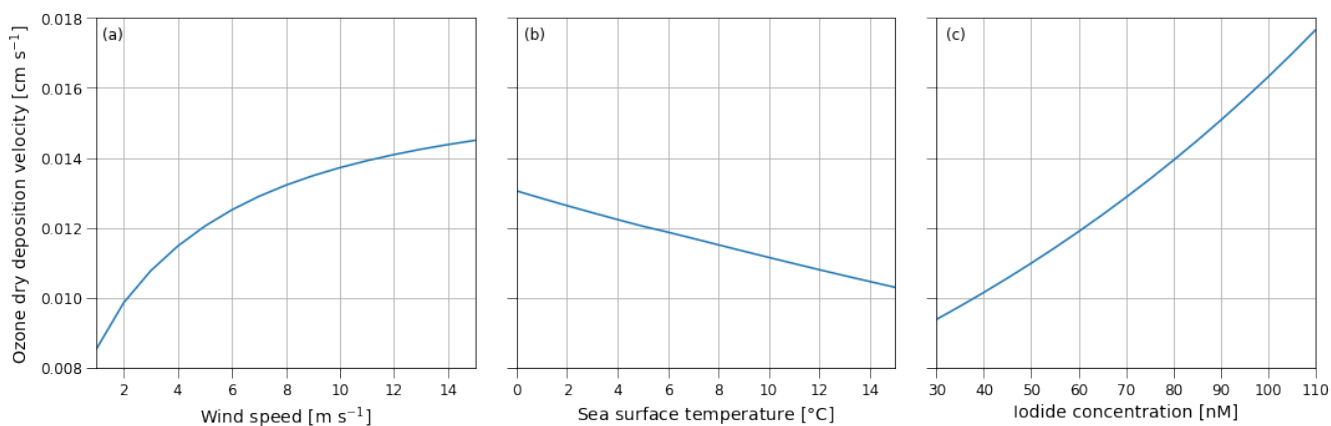


Figure B1. Sensitivity of the ozone dry deposition velocity from COAREG to the environmental factors 10-meter wind speed [m s^{-1}] (a), sea surface temperature [$^\circ\text{C}$] (b) and sea surface Iodide concentration [nM] (c) using typical values of 10-meter wind speed, sea surface temperature and Iodide concentration of 5 m s^{-1} , 5°C and 60 nM respectively. Note that the sensitivity to sea surface temperature does not include effects of increasing reactivity but mostly represents the effect of reduced solubility (Eq. B2).

695

Appendix C: Spatial distribution of oceanic Iodide

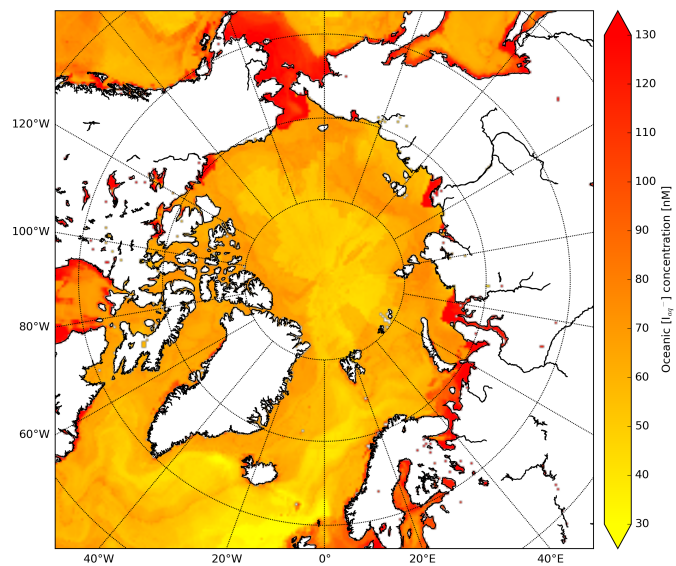


Figure C1. Spatial distribution of Sherwen et al. (2019) oceanic Iodide concentrations [nM] at the start of the simulation.

Appendix D: Surface ozone measurement sites.

Table D1. Surface ozone measurement sites subdivided in the 'High Arctic', 'Remote' and 'Terrestrial' site selections.

Name	Abbreviation	Group	Latitude [°N]	Longitude [°E]
Alert	ALT	High Arctic	82.5	-62.3
ASCOS	ASC	High Arctic	~ 87.4	~ -6.0
Barrow	BRW	High Arctic	71.3	-156.6
Zeppelin	NYA	High Arctic	78.9	11.9
Summit	SUM	High Arctic	72.6	-38.5
Villum	VIL	High Arctic	81.6	-16.7
Denali NP	DEN	Remote	63.7	-149.0
Esrang	ESR	Remote	67.9	21.1
Karasjok	KAS	Remote	69.5	25.2
Inuvik	INU	Remote	68.4	-133.7
Lerwick	SIS	Remote	60.1	-1.2
Pallas	PAL	Remote	68.0	21.1
Storhofdi	ICE	Remote	63.4	-20.3
Yellowknife	YEL	Remote	62.5	-114.4
Ahtari	AHT	Terrestrial	62.6	24.2
Bredkalen	BRE	Terrestrial	63.9	15.3
Fort Liard	FOR	Terrestrial	60.2	-123.5
Hurdal	HUR	Terrestrial	60.4	11.1
Karvatn	KRV	Terrestrial	62.8	8.9
Norman Wells	NOR	Terrestrial	65.3	-123.8
Oulanka	OUX	Terrestrial	66.3	29.4
Tustervatn	TUV	Terrestrial	65.8	13.9
Vindeln	VDI	Terrestrial	64.3	19.8
Virolahti	VIR	Terrestrial	60.5	27.7
Whitehorse	WHI	Terrestrial	60.7	-135.0

References

- Ainsworth, E. A., Yendrek, C. R., Sitch, S., Collins, W. J., and Emberson, L. D.: The effects of tropospheric ozone on net primary productivity and implications for climate change, *Annual review of plant biology*, 63, 637–661, 2012.
- Arnold, S. R., Law, K. S., Brock, C. A., Thomas, J. L., Starkweather, S. M., von Salzen, K., Stohl, A., Sharma, S., Lund, M. T., Flanner, M. G., et al.: Arctic air pollution: Challenges and opportunities for the next decade, *Elementa: Science of the Anthropocene*, 2016.
- Bariteau, L., Helmig, D., Fairall, C., Hare, J., Hueber, J., and Lang, E.: Determination of oceanic ozone deposition by ship-borne eddy covariance flux measurements, *Atmospheric Measurement Techniques*, 3, 441–455, 2010.
- Bell, T. G., Landwehr, S., Miller, S. D., De Bruyn, W. J., Callaghan, A. H., Scanlon, B., Ward, B., Yang, M., and Saltzman, E. S.: Estimation of bubble-mediated air-sea gas exchange from concurrent DMS and CO₂ transfer velocities at intermediate-high wind speeds, *Atmospheric Chemistry and Physics*, 17, 9019–9033, 2017.
- Blomquist, B., Brumer, S., Fairall, C., Huebert, B., Zappa, C., Brooks, I., Yang, M., Bariteau, L., Prytherch, J., Hare, J., et al.: Wind speed and sea state dependencies of air-sea gas transfer: Results from the high wind speed gas exchange study (HiWinGS), *Journal of Geophysical Research: Oceans*, 122, 8034–8062, 2017.
- Bromwich, D. H., Otieno, F. O., Hines, K. M., Manning, K. W., and Shilo, E.: Comprehensive evaluation of polar weather research and forecasting model performance in the Antarctic, *Journal of Geophysical Research: Atmospheres*, 118, 274–292, 2013.
- Chance, R., Baker, A. R., Carpenter, L., and Jickells, T. D.: The distribution of iodide at the sea surface, *Environmental Science: Processes & Impacts*, 16, 1841–1859, 2014.
- Chang, W., Heikes, B. G., and Lee, M.: Ozone deposition to the sea surface: chemical enhancement and wind speed dependence, *Atmospheric Environment*, 38, 1053–1059, 2004.
- Chen, F. and Dudhia, J.: Coupling an advanced land surface–hydrology model with the Penn State–NCAR MM5 modeling system. Part I: Model implementation and sensitivity, *Monthly weather review*, 129, 569–585, 2001.
- Chen, X., Quéléver, L. L., Fung, P. L., Kesti, J., Rissanen, M. P., Bäck, J., Keronen, P., Junninen, H., Petäjä, T., Kerminen, V.-M., et al.: Observations of ozone depletion events in a Finnish boreal forest, *Atmospheric Chemistry and Physics*, 18, 49–63, 2018.
- Clifford, D., Donaldson, D., Brigante, M., D’Anna, B., and George, C.: Reactive uptake of ozone by chlorophyll at aqueous surfaces, *Environmental science & technology*, 42, 1138–1143, 2008.
- Clifton, O., Paulot, F., Fiore, A., Horowitz, L., Correa, G., Baublitz, C., Fares, S., Goded, I., Goldstein, A., Gruening, C., et al.: Influence of dynamic ozone dry deposition on ozone pollution, *Journal of Geophysical Research: Atmospheres*, 125, e2020JD032 398, 2020a.
- Clifton, O. E., Fiore, A. M., Massman, W. J., Baublitz, C. B., Coyle, M., Emberson, L., Fares, S., Farmer, D. K., Gentine, P., Gerosa, G., et al.: Dry deposition of ozone over land: processes, measurement, and modeling, *Reviews of Geophysics*, 58, e2019RG000 670, 2020b.
- Cooper, O. R., Parrish, D., Ziemke, J., Cupeiro, M., Galbally, I., Gilge, S., Horowitz, L., Jensen, N., Lamarque, J.-F., Naik, V., et al.: Global distribution and trends of tropospheric ozone: An observation-based review, 2014.
- Cooper, O. R., Schultz, M. G., Schröder, S., Chang, K.-L., Gaudel, A., Benítez, G. C., Cuevas, E., Fröhlich, M., Galbally, I. E., Molloy, S., et al.: Multi-decadal surface ozone trends at globally distributed remote locations, *Elementa: Science of the Anthropocene*, 8, 2020.
- Dee, D. P., Uppala, S. M., Simmons, A., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M., Balsamo, G., Bauer, d. P., et al.: The ERA-Interim reanalysis: Configuration and performance of the data assimilation system, *Quarterly Journal of the royal meteorological society*, 137, 553–597, 2011.

- Fairall, C., Helmig, D., Ganzeveld, L., and Hare, J.: Water-side turbulence enhancement of ozone deposition to the ocean, *Atmospheric Chemistry and Physics*, 2007, 443–451, 2007.
- 735 Fairall, C., Yang, M., Bariteau, L., Edson, J., Helmig, D., McGillis, W., Pezoa, S., Hare, J., Huebert, B., and Blomquist, B.: Implementation of the Coupled Ocean-Atmosphere Response Experiment flux algorithm with CO₂, dimethyl sulfide, and O₃, *Journal of Geophysical Research: Oceans*, 116, 2011.
- Fairall, C. W., Bradley, E. F., Rogers, D. P., Edson, J. B., and Young, G. S.: Bulk parameterization of air-sea fluxes for tropical ocean-global atmosphere coupled-ocean atmosphere response experiment, *Journal of Geophysical Research: Oceans*, 101, 3747–3764, 1996.
- 740 Fan, S.-M., Wofsy, S. C., Bakwin, P. S., Jacob, D. J., and Fitzjarrald, D. R.: Atmosphere-biosphere exchange of CO₂ and O₃ in the central Amazon forest, *Journal of Geophysical Research: Atmospheres*, 95, 16 851–16 864, 1990.
- Gallagher, M., Beswick, K., and Coe, H.: Ozone deposition to coastal waters, *Quarterly Journal of the Royal Meteorological Society*, 127, 539–558, 2001.
- 745 Ganzeveld, L., Helmig, D., Fairall, C., Hare, J., and Pozzer, A.: Atmosphere-ocean ozone exchange: A global modeling study of biogeochemical, atmospheric, and waterside turbulence dependencies, *Global Biogeochemical Cycles*, 23, 2009.
- Gaudel, A., Cooper, O. R., Chang, K.-L., Bourgeois, I., Ziemke, J. R., Strode, S. A., Oman, L. D., Sellitto, P., Nédélec, P., Blot, R., et al.: Aircraft observations since the 1990s reveal increases of tropospheric ozone at multiple locations across the Northern Hemisphere, *Science Advances*, 6, eaba8272, 2020.
- 750 Gery, M. W., Whitten, G. Z., Killus, J. P., and Dodge, M. C.: A photochemical kinetics mechanism for urban and regional scale computer modeling, *Journal of Geophysical Research: Atmospheres*, 94, 12 925–12 956, 1989.
- Gorter, W., Van Angelen, J., Lenaerts, J., and Van den Broeke, M.: Present and future near-surface wind climate of Greenland from high resolution regional climate modelling, *Climate dynamics*, 42, 1595–1611, 2014.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled “online” chemistry within the WRF model, *Atmospheric Environment*, 39, 6957–6975, 2005.
- 755 Guenther, A., Jiang, X., Heald, C., Sakulyanontvittaya, T., Duhl, T., Emmons, L., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2. 1): an extended and updated framework for modeling biogenic emissions, 2012.
- Halfacre, J., Knepp, T., Shepson, P., Thompson, C., Pratt, K., Li, B., Peterson, P., Walsh, S., Simpson, W., Matrai, P., et al.: Temporal and spatial characteristics of ozone depletion events from measurements in the Arctic, *Atmospheric Chemistry and Physics*, 14, 4875, 2014.
- 760 Hardacre, C., Wild, O., and Emberson, L.: An evaluation of ozone dry deposition in global scale chemistry climate models, *Atmospheric Chemistry and Physics*, 15, 6419–6436, 2015.
- Helmig, D., Ganzeveld, L., Butler, T., and Oltmans, S.: The role of ozone atmosphere-snow gas exchange on polar, boundary-layer tropospheric ozone? a review and sensitivity analysis, 2007a.
- Helmig, D., Oltmans, S. J., Carlson, D., Lamarque, J.-F., Jones, A., Labuschagne, C., Anlauf, K., and Hayden, K.: A review of surface ozone in the polar regions, *Atmospheric Environment*, 41, 5138–5161, 2007b.
- 765 Helmig, D., Cohen, L. D., Bocquet, F., Oltmans, S., Grachev, A., and Neff, W.: Spring and summertime diurnal surface ozone fluxes over the polar snow at Summit, Greenland, *Geophysical research letters*, 36, 2009.
- Helmig, D., Lang, E., Bariteau, L., Boylan, P., Fairall, C., Ganzeveld, L., Hare, J., Hueber, J., and Pallandt, M.: Atmosphere-ocean ozone fluxes during the TexAQS 2006, STRATUS 2006, GOMECC 2007, GasEx 2008, and AMMA 2008 cruises, *Journal of Geophysical Research: Atmospheres*, 117, 2012.
- 770

- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., et al.: The ERA5 global reanalysis, *Quarterly Journal of the Royal Meteorological Society*, 2020.
- Hines, K. M. and Bromwich, D. H.: Development and testing of Polar Weather Research and Forecasting (WRF) model. Part I: Greenland ice sheet meteorology, *Monthly Weather Review*, 136, 1971–1989, 2008.
- 775 Hong, S.-Y., Dudhia, J., and Chen, S.-H.: A revised approach to ice microphysical processes for the bulk parameterization of clouds and precipitation, *Monthly weather review*, 132, 103–120, 2004.
- Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and Collins, W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models, *Journal of Geophysical Research: Atmospheres*, 113, 2008.
- Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, A.-M., Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling, Z., Massart, S., Parrington, M., Peuch, V.-H., Razinger, M., Remy, S., Schulz, M., and Suttie, M.: 780 The CAMS reanalysis of atmospheric composition, *Atmospheric Chemistry and Physics*, 19, 3515–3556, <https://doi.org/10.5194/acp-19-3515-2019>, <https://www.atmos-chem-phys.net/19/3515/2019/>, 2019.
- Janjić, Z. I.: The step-mountain eta coordinate model: Further developments of the convection, viscous sublayer, and turbulence closure schemes, *Monthly weather review*, 122, 927–945, 1994.
- 785 Janjić, Z. I.: Nonsingular implementation of the Mellor-Yamada level 2.5 scheme in the NCEP Meso model, 2001.
- Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., Bergamaschi, P., Pagliari, V., Olivier, J., Peters, J., et al.: EDGAR v4.3.2 Global Atlas of the three major Greenhouse Gas Emissions for the period 1970–2012, *Earth Syst. Sci. Data Discuss*, 2017.
- Kain, J. S.: The Kain-Fritsch convective parameterization: an update, *Journal of applied meteorology*, 43, 170–181, 2004.
- 790 Kavassalis, S. C. and Murphy, J. G.: Understanding ozone-meteorology correlations: A role for dry deposition, *Geophysical Research Letters*, 44, 2922–2931, 2017.
- Klein, T., Heinemann, G., Bromwich, D. H., Cassano, J. J., and Hines, K. M.: Mesoscale modeling of katabatic winds over Greenland and comparisons with AWS and aircraft data, *Meteorology and Atmospheric Physics*, 78, 115–132, 2001.
- Lana, A., Bell, T., Simó, R., Vallina, S., Ballabrera-Poy, J., Kettle, A., Dachs, J., Bopp, L., Saltzman, E., Stefels, J., et al.: An updated 795 climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean, *Global Biogeochemical Cycles*, 25, 2011.
- Law, K. S., Roiger, A., Thomas, J. L., Marelle, L., Raut, J.-C., Dalsøren, S., Fuglestedt, J., Tuccella, P., Weinzierl, B., and Schlager, H.: Local Arctic air pollution: Sources and impacts, *Ambio*, 46, 453–463, 2017.
- Lee, D. S., Holland, M. R., and Falla, N.: The potential impact of ozone on materials in the UK, *Atmospheric Environment*, 30, 1053–1065, 1996.
- 800 Lelieveld, J. and Dentener, F. J.: What controls tropospheric ozone?, *Journal of Geophysical Research: Atmospheres*, 105, 3531–3551, 2000.
- Lin, M., Horowitz, L. W., Payton, R., Fiore, A. M., and Tonnesen, G.: US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, *Atmospheric Chemistry and Physics*, 17, 2943–2970, 2017.
- Lin, M., Malyshev, S., Shevliakova, E., Paulot, F., Horowitz, L. W., Fares, S., Mikkelsen, T. N., and Zhang, L.: Sensitivity of ozone dry deposition to ecosystem-atmosphere interactions: A critical appraisal of observations and simulations, *Global Biogeochemical Cycles*, 33, 805 1264–1288, 2019.
- Lin, M., Horowitz, L. W., Xie, Y., Paulot, F., Malyshev, S., Shevliakova, E., Finco, A., Gerosa, G., Kubistin, D., and Pilegaard, K.: Vegetation feedbacks during drought exacerbate ozone air pollution extremes in Europe, *Nature Climate Change*, 10, 444–451, 2020.

- Luhar, A. K., Galbally, I. E., Woodhouse, M. T., and Thatcher, M.: An improved parameterisation of ozone dry deposition to the ocean and its impact in a global climate-chemistry model, *Atmospheric Chemistry and Physics*, 17, 3749, 2017.
- 810 Luhar, A. K., Woodhouse, M. T., and Galbally, I. E.: A revised global ozone dry deposition estimate based on a new two-layer parameterisation for air-sea exchange and the multi-year MACC composition reanalysis., *Atmospheric Chemistry & Physics*, 18, 2018.
- MacDonald, S., Gómez Martín, J., Chance, R., Warriner, S., Saiz-Lopez, A., Carpenter, L., and Plane, J.: A laboratory characterisation of inorganic iodine emissions from the sea surface: dependence on oceanic variables and parameterisation for global modelling, *Atmospheric Chemistry and Physics*, 14, 5841–5852, 2014.
- 815 Magi, L., Schweitzer, F., Pallares, C., Cherif, S., Mirabel, P., and George, C.: Investigation of the uptake rate of ozone and methyl hydroperoxide by water surfaces, *The Journal of Physical Chemistry A*, 101, 4943–4949, 1997.
- Mahmood, R., von Salzen, K., Norman, A.-L., Galí, M., and Levasseur, M.: Sensitivity of Arctic sulfate aerosol and clouds to changes in future surface seawater dimethylsulfide concentrations., *Atmospheric Chemistry & Physics*, 19, 2019.
- Marelle, L., Thomas, J. L., Raut, J.-C., Law, K. S., Jalkanen, J.-P., Johansson, L., Roiger, A., Schlager, H., Kim, J., Reiter, A., et al.: Air
820 quality and radiative impacts of Arctic shipping emissions in the summertime in northern Norway: from the local to the regional scale, 2016.
- Marelle, L., Raut, J.-C., Law, K. S., Berg, L. K., Fast, J. D., Easter, R. C., Shrivastava, M., and Thomas, J. L.: Improvements to the WRF-Chem 3.5.1 model for quasi-hemispheric simulations of aerosols and ozone in the Arctic, *Geoscientific Model Development*, 10, 3661–3677, <https://doi.org/10.5194/gmd-10-3661-2017>, <https://gmd.copernicus.org/articles/10/3661/2017/>, 2017.
- 825 Marelle, L., Raut, J.-C., Law, K. S., and Duclaux, O.: Current and Future Arctic Aerosols and Ozone From Remote Emissions and Emerging Local Sources—Modeled Source Contributions and Radiative Effects, *Journal of Geophysical Research: Atmospheres*, 123, 12–942, 2018.
- Martino, M., Lézé, B., Baker, A. R., and Liss, P. S.: Chemical controls on ozone deposition to water, *Geophysical research letters*, 39, 2012.
- Michou, M., Laville, P., Serça, D., Fotiadi, A., Bouchou, P., and Peuch, V.-H.: Measured and modeled dry deposition velocities over the ESCOMPTE area, *Atmospheric Research*, 74, 89–116, 2005.
- 830 Morris, J.: The aqueous solubility of ozone—A review, *Ozone news*, 1, 14–16, 1988.
- Muller, J. B., Dorsey, J. R., Flynn, M., Gallagher, M. W., Percival, C. J., Shallcross, D. E., Archibald, A., Roscoe, H. K., Obbard, R. W., Atkinson, H. M., et al.: Energy and ozone fluxes over sea ice, *Atmospheric environment*, 47, 218–225, 2012.
- Murray, K. A., Kramer, L. J., Doskey, P. V., Ganzeveld, L., Seok, B., Van Dam, B., and Helmig, D.: Dynamics of ozone and nitrogen oxides at Summit, Greenland. II. Simulating snowpack chemistry during a spring high ozone event with a 1-D process-scale model, *Atmospheric
835 Environment*, 117, 110–123, 2015.
- Nguyen, Q. T., Glasius, M., Sørensen, L. L., Jensen, B., Skov, H., Birmili, W., Wiedensohler, A., Kristensson, A., Nøjgaard, J. K., and Massling, A.: Seasonal variation of atmospheric particle number concentrations, new particle formation and atmospheric oxidation capacity at the high Arctic site Villum Research Station, Station Nord, 2016.
- Nuvolone, D., Petri, D., and Voller, F.: The effects of ozone on human health, *Environmental Science and Pollution Research*, 25, 8074–8088,
840 2018.
- Oh, I.-B., Byun, D. W., Kim, H.-C., Kim, S., and Cameron, B.: Modeling the effect of iodide distribution on ozone deposition to seawater surface, *Atmospheric Environment*, 42, 4453–4466, 2008.
- Oltmans, S., Lefohn, A., Shadwick, D., Harris, J., Scheel, H., Galbally, I., Tarasick, D., Johnson, B., Brunke, E.-G., Claude, H., et al.: Recent tropospheric ozone changes—A pattern dominated by slow or no growth, *Atmospheric Environment*, 67, 331–351, 2013.

- 845 Paatero, J., Vaattovaara, P., Vestenius, M., Meinander, O., Makkonen, U., Kivi, R., Hyvärinen, A., Asmi, E., Tjernström, M., and Leck, C.: Finnish contribution to the arctic summer cloud ocean study (ASCOS) expedition, *Arctic Ocean 2008*, *Geophysica*, 45, 119–146, 2009.
- Padro, J.: Summary of ozone dry deposition velocity measurements and model estimates over vineyard, cotton, grass and deciduous forest in summer, *Atmospheric Environment*, 30, 2363–2369, 1996.
- Pausata, F., Pozzoli, L., Vignati, E., and Dentener, F.: North Atlantic Oscillation and tropospheric ozone variability in Europe: model analysis
850 and measurements intercomparison., *Atmospheric Chemistry & Physics*, 12, 2012.
- Pedregosa, F., Varoquaux, G., Gramfort, A., Michel, V., Thirion, B., Grisel, O., Blondel, M., Prettenhofer, P., Weiss, R., Dubourg, V., et al.: Scikit-learn: Machine learning in Python, *the Journal of machine Learning research*, 12, 2825–2830, 2011.
- Porter, J., de Bruyn, W., Miller, S., and Saltzman, E.: Air/sea transfer of highly soluble gases over coastal waters, *Geophysical Research Letters*, 47, no–no, 2020.
- 855 Pound, R. J., Sherwen, T., Helmig, D., Carpenter, L. J., and Evans, M. J.: Influences of oceanic ozone deposition on tropospheric photochemistry, *Atmospheric Chemistry and Physics Discussions*, pp. 1–25, 2019.
- Prados Roman, C., Cuevas, C. A., Fernandez, R. P., Kinnison, D. E., Lamarque, J. F., and Saiz-lopez, A.: A negative feedback between anthropogenic ozone pollution and enhanced ocean emissions of iodine, 2015.
- Pratt, K. A., Custard, K. D., Shepson, P. B., Douglas, T. A., Pöhler, D., General, S., Zielcke, J., Simpson, W. R., Platt, U., Tanner, D. J., et al.:
860 Photochemical production of molecular bromine in Arctic surface snowpacks, *Nature Geoscience*, 6, 351–356, 2013.
- Reeser, D. I., Jammoul, A., Clifford, D., Brigante, M., D’Anna, B., George, C., and Donaldson, D.: Photoenhanced reaction of ozone with chlorophyll at the seawater surface, *The Journal of Physical Chemistry C*, 113, 2071–2077, 2009.
- Riedel, A., Michel, C., Gosselin, M., and LeBlanc, B.: Winter–spring dynamics in sea-ice carbon cycling in the coastal Arctic Ocean, *Journal of Marine Systems*, 74, 918–932, 2008.
- 865 Schmale, J., Arnold, S., Law, K. S., Thorp, T., Anenberg, S., Simpson, W., Mao, J., and Pratt, K.: Local Arctic air pollution: A neglected but serious problem, *Earth’s Future*, 6, 1385–1412, 2018.
- Sherwen, T., Chance, R. J., Tinel, L., Ellis, D., Evans, M. J., and Carpenter, L. J.: A machine learning based global sea-surface iodide distribution, *Earth System Science Data Discussions*, pp. 1–40, 2019.
- Silva, S. J. and Heald, C. L.: Investigating dry deposition of ozone to vegetation, *Journal of Geophysical Research: Atmospheres*, 123,
870 559–573, 2018.
- Stefels, J., Steinke, M., Turner, S., Malin, G., and Belviso, S.: Environmental constraints on the production and removal of the climatically active gas dimethylsulphide (DMS) and implications for ecosystem modelling, *Biogeochemistry*, 83, 245–275, 2007.
- Tarasick, D., Galbally, I. E., Cooper, O. R., Schultz, M. G., Ancellet, G., Leblanc, T., Wallington, T. J., Ziemke, J., Liu, X., Steinbacher, M., et al.: Tropospheric Ozone Assessment Report: Tropospheric ozone from 1877 to 2016, observed levels, trends and uncertainties,
875 *Elementa: Science of the Anthropocene*, 7, 2019.
- Thomas, J. L., Stutz, J., Lefer, B., Huey, L. G., Toyota, K., Dibb, J. E., and von Glasow, R.: Modeling chemistry in and above snow at Summit, Greenland – Part 1: Model description and results, *Atmospheric Chemistry and Physics*, 11, 4899–4914, <https://doi.org/10.5194/acp-11-4899-2011>, <https://acp.copernicus.org/articles/11/4899/2011/>, 2011.
- Thomas, J. L., Raut, J.-C., Law, K. S., Marelle, L., Ancellet, G., Ravetta, F., Fast, J. D., Pfister, G., Emmons, L. K., Diskin, G. S., Weinheimer, A., Roiger, A., and Schlager, H.: Pollution transport from North America to Greenland during summer 2008, *Atmospheric Chemistry and Physics*, 13, 3825–3848, <https://doi.org/10.5194/acp-13-3825-2013>, <https://www.atmos-chem-phys.net/13/3825/2013/>, 2013.

- Thompson, C. R., Shepson, P. B., Liao, J., Huey, L. G., Cantrell, C., Flocke, F., and Orlando, J.: Bromine atom production and chain propagation during springtime Arctic ozone depletion events in Barrow, Alaska, *Atmospheric Chemistry and Physics*, 17, 3401, 2017.
- 885 Tjernstrom, M., Birch, C. E., Brooks, I. M., Shupe, M. D., Persson, P. O. G., Sedlar, J., Mauritsen, T., Leck, C., Paatero, J., Szczodrak, M., et al.: Meteorological conditions in the central Arctic summer during the Arctic Summer Cloud Ocean Study (ASCOS), *Atmospheric Chemistry and Physics*, 12, 6863–6889, 2012.
- Toyota, K., McConnell, J., Staebler, R., and Dastoor, A.: Air–snowpack exchange of bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS–Part 1: In-snow bromine activation and its impact on ozone, *Atmos. Chem. Phys.*, 14, 4101–4133, 2014.
- 890 Toyota, K., Dastoor, A. P., and Ryzhkov, A.: Parameterization of gaseous dry deposition in atmospheric chemistry models: Sensitivity to aerodynamic resistance formulations under statically stable conditions, *Atmospheric Environment*, 147, 409–422, 2016.
- Val Martin, M., Heald, C., and Arnold, S.: Coupling dry deposition to vegetation phenology in the Community Earth System Model: Implications for the simulation of surface O₃, *Geophysical Research Letters*, 41, 2988–2996, 2014.
- Van Dam, B., Helmig, D., Toro, C., Doskey, P., Kramer, L., Murray, K., Ganzeveld, L., and Seok, B.: Dynamics of ozone and nitrogen oxides at Summit, Greenland: I. Multi-year observations in the snowpack, *Atmospheric Environment*, 123, 268–284, 2015.
- 895 Van Dam, B., Helmig, D., Doskey, P. V., and Oltmans, S. J.: Summertime surface O₃ behavior and deposition to tundra in the Alaskan Arctic, *Journal of Geophysical Research: Atmospheres*, 121, 8055–8066, 2016.
- Wentz, F. and Meissner, T.: AMSR-E/Aqua Daily L3 Global Ascending/Descending .25x.25 deg Ocean Grids, Version 2., 2004.
- Wesely, M.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, *Atmospheric Environment* 900 (1967), 23, 1293–1304, 1989.
- Wesely, M. and Hicks, B.: A review of the current status of knowledge on dry deposition, *Atmospheric environment*, 34, 2261–2282, 2000.
- Wild, O., Zhu, X., and Prather, M. J.: Fast-J: Accurate simulation of in-and below-cloud photolysis in tropospheric chemical models, *Journal of Atmospheric Chemistry*, 37, 245–282, 2000.
- Wong, A. Y., Geddes, J. A., Tai, A. P., and Silva, S. J.: Importance of dry deposition parameterization choice in global simulations of surface 905 ozone, *Atmospheric Chemistry and Physics*, 19, 14 365–14 385, 2019.
- Yang, X., Blechschmidt, A.-M., Bogner, K., McClure-Begley, A., Morris, S., Petropavlovskikh, I., Richter, A., Skov, H., Strong, K., Tarasick, D. W., et al.: Pan-Arctic surface ozone: modelling vs. measurements, *Atmospheric Chemistry and Physics*, 20, 15 937–15 967, 2020.
- Young, P. J., Naik, V., Fiore, A. M., Gaudel, A., Guo, J., Lin, M., Neu, J., Parrish, D., Rieder, H., Schnell, J., et al.: Tropospheric Ozone Assessment Report: Assessment of global-scale model performance for global and regional ozone distributions, variability, and trends, 910 *Elementa: Science of the Anthropocene*, 6, 2018.
- Zaveri, R. A. and Peters, L. K.: A new lumped structure photochemical mechanism for large-scale applications, *Journal of Geophysical Research: Atmospheres*, 104, 30 387–30 415, 1999.
- Zeller, K.: Wintertime ozone fluxes and profiles above a subalpine spruce–fir forest, *Journal of Applied Meteorology*, 39, 92–101, 2000.
- Zhou, P., Ganzeveld, L., Rannik, Ü., Zhou, L., Gierens, R., Taipale, D., Mammarella, I., and Boy, M.: Simulating ozone dry deposition at a 915 boreal forest with a multi-layer canopy deposition model, *Atmospheric Chemistry and Physics*, 17, 1361–1379, 2017.