Author Responses to RC3: 'Review of "Simulation study for ground-based Ku-band microwave observations of ozone and hydroxyl in the polar middle atmosphere" by Newnham et al.', Anonymous Referee #1

We provide a point-by-point response to the review by Anonymous Referee #1. Each comment is reproduced in italics followed by our response and the changes made in the manuscript, with the marked-up manuscript version appended. The line numbers identifying changes refer to the marked-up manuscript.

Author response to Anonymous Referee #1

We thank Anonymous Referee #1 for their comments (*Atmos. Meas. Tech. Discuss.,* doi:10.5194/amt-2018-334-RC3, 2018; https://www.atmos-meas-tech-discuss.net/amt-2018-334/amt-2018-334-RC3-supplement.pdf) on our manuscript. In particular we welcome their suggested changes aimed at improving the paper. The referee states that the study is worth publishing in AMT after considering their points. Our response, and proposed changes to the manuscript to address the points raised in their review, are given below.

General comments

Since this study is purely theoretical, no actual measurements can be presented. However, the authors fall short on describing how feasible actual measurements are in the proposed way. That is, are there any cost limitations and how hard would it be to combine the signals from 40 receivers (for OH) or to cool them to 46 K? Are there any plans to actually carry out such measurements?

Further details have been added in the Conclusions section (Page 13, lines 3–28) on the feasibility and merits of making the proposed measurements either by combining the signals from multiple receivers or using a single cooled receiver, as well as the next steps towards building and operating an actual instrument. References for three new citations (de Waele, 2011; Radebaugh, 2009; Tenneti and Rogers, 2009) have been added to the References section (Page 15, lines 18–19; Page 16, lines 29–30; Page 18, lines 1–2).

Subsection 2.1 describes the model atmosphere and its variation during a sub-storm to great length. This model atmosphere was used to evaluate the retrievals but the authors fail to properly link the rather extensively described ozone and OH model variations to their impact on the retrieval. Does it matter for the current study that "the largest decrease in O_3 occurred between 01:30–07:30 UTC on day 363"? How typical are these values and variations to occur in the real world, are they representative for the general case? All those timing and the discussion can be removed, and subsection 2.1 can be shortened substantially by concentrating on the values that are important for the retrieval simulations. The authors should then provide a better logical connection to the rest of the manuscript, i.e. to the retrieval simulations.

The detailed timings of the largest decrease in O_3 and the largest increase in OH have been removed as suggested (Page 5, lines 14–15 and 27–28). However, we disagree with the referee's suggestion that all of the discussion can be removed. Subsection 2.1 describes how the atmospheric profiles used in the retrieval simulations are derived from the original SIC model and WACCM-D datasets. In the absence of suitable current observations, these model data are the best existing representation of the short-term variations in mesospheric O_3 and OH due to energetic electron precipitation. Furthermore, the description explains how the associated data-sets (Model data for simulating atmospheric microwave spectra at 11.072 GHz and 13.441 GHz and performing retrievals of ozone (O_3) and hydroxyl (OH) vertical profiles, D. Newnham, P. Verronen, and A. Seppälä, https://doi.org/10.5285/57858a9a-d814-412c-8e79-9a542cd055d4) are derived. We therefore wish to keep the remaining original description of the atmospheric data-sets.

Minor points and detailed comments

Page 2, line 11: Relative increases of a relative quantity can be misleading and OH has a large diurnal variability. Further, a relevant study should be cited that derives this 1000% increase.

The sentence describing the modelled increases in OH abundances has been clarified to read as follows (Page 2, lines 12–14):-
"During the sub storms the modelled night-time OH partial column over the altitude range 64–84 km can increase by more than 1000% and the peak in OH volume mixing ratio (VMR) at 70 km increases from the background (i.e. no sub storm) level of ~8 ppbv to ~60 ppbv (Seppälä et al., 2015)."

Page 2, line 12: Please provide a reference for the 5–55% ozone loss.

A reference (Seppälä et al., 2015) for the 5–55% ozone loss has been added (Page 2, line 16).

Page 2, line 25: Is "much more" really needed when "abundant" is used? I'd suggest to change to something like "... abundant during night and depleted during day ...".

The suggested change has been made (Page 2, line 28).

Page 2, line 27: "Another instrument" should be removed and the commas adjusted accordingly.

The suggested change has been made (Page 2, line 30).

Page 2, line 30: I'd suggest to change to: "helped to elucidate".

The suggested change has been made (Page 2, line 33).

Page 3, line 32: Insert "the" before "Λ-doubling hyperfine structure".

The suggested change has been made (Page 4, line 2).

Page 4, line 17: I'd suggest to change to: "are given in Section 2.3."

The suggested change has been made (Page 4, line 20).

Page 4, line 20: I'd suggest to remove the hyphen in front of "ozone".

The suggested change has been made (Page 4, line 23).

Page 4, line 23: I'd suggest to move "from WACCM-D" to before "covering 2000–2009".

The suggested change has been made (Page 4, lines 26–27).

Page 4, line 26: There is something missing, it does not seem to be a complete sentence. Perhaps: "The model data were taken from the ... points closest to ...".

The suggested change has been made (Page 4, line 29).

Page 5, line 4: I'd suggest to swap "during a four-day period" and "over the altitude range 68–86 km".

The suggested change has been made (Page 5, lines 8–9).

Page 5, line 5: Insert "the" in front of "number densities".

The requested change has been made (Page 5, line 9).

Page 5, line 12: How large is the range around 75 km?

The sentence has been changed (Page 5, lines 16–17) to:-

"Sub-storm reductions in O_3 VMR occur over the altitude range 64–94 km, with the largest decreases at 72 km."

Page 5, lines 12–14: This is a long and a little convoluted sentence which is hard to understand. Why are the SIC ozone values larger than the SIC ozone values? However, since this discussion seems to be unimportant for the rest of the study, it could just be removed.

The sentences describing Fig 2 have been revised (Page 5, lines 12–16) and also the corresponding text describing Fig 3 (Page 5, lines 28–30). We agree that discussion of differences between the SIC model ozone profiles and the monthly-mean WACCM-D data is unimportant here and, as suggested, this has been removed (Page 5, lines 17–19).

Page 6, line 1: I'd suggest to insert "taken" after "were": ". . . were taken from ...".

The suggested change has been made (Page 6, line 9).

Page 6, lines 4–5: I'd suggest to begin the sentence with "As the water vapour . . . ", to remove "used was", and add "was used" after the model version number before the comma.

The suggested change has been made (Page 6, lines 12–13).

Page 6, line 5: "separately" can probably be removed.

The suggested change has been made (Page 6, line 13).

Page 6, line 7: In my opinion "therefore" would fit a little better than "hence" here, but I leave that decision to the authors.

The suggested change has been made (Page 6, line 15).

Page 7, line 16: Insert "taken" after "were": "... were taken from ... ". Why was the older version IGRF-11 used and not the IGRF-12?

The suggested change has been made (Page 7, line 25). The ARTS model used in the study was already setup to use IGRF-11 inputs. For this proof-of-concept feasibility study we considered it unimportant to spend time changing the code to use the more up-to-date IGRF-12. For analysing actual observations of Zeeman-split OH lines it would be important to use the most up-to-date and accurate representation of the Earth's magnetic field along the observing path from IGRF or local magnetometer observations.

Page 7, line 16: What does "renormalized" mean? I'd suggest to use "normalized" to avoid confusion with renormalization as it is known in theoretical physics, e.g. in QED.

In order to avoid confusion the word "renormalized" has been removed so the sentence now reads (Page 7, lines 25–26):-

"… and the Faddeeva function was used to describe the lineshape."

Page 7, line 24: What distribution was used for the scaling factor? Was it uniform or a normal distribution around 1? The distribution of the scaling factor may have an impact on how useful the resulting distribution is for evaluating the uncertainties in the retrieval algorithm. Here the authors need to be more specific and more careful about the sampled distribution.

The scaling factors were calculated using a uniform distribution over the range 0.5–2.0. We are confident that this distribution provides a suitable range of O_3, OH, and H_2O VMR profiles to assess the performance of the retrieval algorithm. The sentence has been changed to the type of distribution used (Page 8, line 1):-

"… were randomly scaled using a uniform distribution over the range 0.5–2.0."

Page 7, line 29: I'd suggest to use "within" instead of "inside".

The suggested change has been made (Page 8, line 6).

Page 7, line 31: I'd suggest to use "for" instead of "over".

The suggested change has been made (Page 8, line 8).

Page 7, line 32: To keep the tenses consistent, "is assumed" should probably be "was assumed".

The suggested change has been made (Page 8, line 9).

Page 7, line 33: I'd suggest to change to: ". . . fixed to 1.5 ppmv ...".

The suggested change has been made (Page 8, line 10).

Page 8, lines 1–3: This description is rather convolved and confusing. I'd suggest to restructure: "The off-diagonal elements

of ... linearly decrease with a correlation length of one fifth ...".

The suggested change has been made (Page 8, lines 12–13).

Page 8, line 10: I'd suggest to change to "At a zenith angle of 82° the lower transmittance ..."

The suggested change has been made (Page 8, line 20).

Page 8, line 12: I'd suggest to change to "... that the different ..."

The suggested change has been made (Page 8, line 22).

Page 8, line 17: I'd suggest to use "at a zenith angle of 82°."

The suggested change has been made (Page 8, line 27).

Page 9, line 15: I cannot find a reference Rodgers, 2004 in the citation list which is presumably a typo and should read

"Rodgers, 2000".

The citation has been corrected to "Rodgers, 2000" (Page 9, line 27).

Page 9, line 18: I'd suggest to remove "amount of".

The suggested change has been made (Page 9, line 30).

Page 9, line 20: I'd suggest to change "estimated" to "identified".

The suggested change has been made (Page 9, line 32).

Page 9, line 20: I'd suggest to insert "altitude" before "range".

The suggested change has been made (Page 9, line 32).

Page 9, line 22: I'd suggest to insert an article "the" before "O_3".

The suggested change has been made (Page 10, line 2).

Page 9, line 23: As it is written now, this sentence suggests that "the retrieved O_3" is responsible for smoothing the information. I'd suggest to rewrite that sentence to state what the finite width of an averaging kernel means.

We agree that the original sentence is ambiguous. It has therefore been amended to read as follows (Page 10, lines 3–5).

"The AVKs indicate the range of altitudes over which O_3 is observed. In the ideal case the AVKs would be delta functions but in practice they are peaked functions with finite widths dependent on the spatial resolution of the observing system."

Page 9, line 24: I'd suggest to remove "Thus," and to start the sentence with "The full-width ... " since "Thus" would need a bit more explanation than simply referring to the smoothing effect of the AVKs. Second, I believe that it should be "widths of the kernels provide" or "width of the kernel provides".

The suggested changes have been made (Page 10, lines 5–6).

Page 9, line 25: In my point of view, if the altitude resolution is changing from 10.9 km to 18.4 km, the resolution gets worse and is therefore decreasing.

The wording has been changed to "altitude resolutions decreasing" (Page 10, line 7).

Page 9, line 29: I'd suggest to introduce a paragraph break before "The OEM ...".

The suggested change has been made (Page 10, lines 11–12).

Page 9, line 30: I'd suggest to change "to give further . . . retrieved profiles." to: "which provide further diagnostic uncertainty estimates of the retrieved profiles."

The suggested change has been made (Page 10, lines 12–13).

Page 9, line 32: Are the stated 0.34 ppmv exact? I'd suggest to insert "around" or "about" before that number.

The wording has been changed to "about 0.34 ppmv" as suggested (Page 10, lines 14–15).

Page 10, line 1: I'd suggest to move "outside the range of the AVK peaks" to the end of that sentence.

The suggested change has been made (Page 10, lines 17–18).

Page 10, line 3: I'd suggest to insert "the" in front of "MC".

The suggested change has been made (Page 11, line 19).

Page 10, lines 3–5: See the point above about the scaling factor distribution used for the MC samples.

The sentence has been changed to state the type of distribution used (Page 10, lines 19–10):-

"… were randomly scaled using a uniform distribution over the range 0.5–2.0."

Page 10, line 8: "are dependent" can be shortened to "depend".

The suggested change has been made (Page 10, line 24).

Page 10, line 10: I'd suggest to change "for" to "of". This sentence is confusing, "should be adequately sampled" by whom? Doesn't the Jacobian depend on the retrieval setup (forward model) and therefore this adequate sampling should rather refer to that?

The suggested change ("for" to "of") has been made (Page 10, line 27). The text has been revised as follows to better describe the sampling and Jacobian for O_3 and, later on, for OH (Page 10, lines 27–30; Page 12, lines 6–9):-

"The Jacobian and gain matrices of the O_3 forward model indicate that an instrument with 10 kHz frequency resolution will adequately sample the emission line for altitudes up to ~98 km where the O_3 measurement contributes significantly to the retrieval. The values of the Jacobian, …".

"The Jacobian and gain matrices of the OH vertical polarisation forward model indicate that, as for O_3, the OH emission line should be adequately sampled by an instrument with 10 kHz frequency resolution for all altitudes where the OH measurement contributes significantly to the retrieval. The values of the Jacobian describing the OH retrieval, …".

Page 10, line 25: I believe it should be "averages and standard deviations" or "average and standard deviation".

The text has been changed to "average and standard deviation" as suggested (Page 11, line 10).

Page 10, line 27: I'd suggest to use "approaches" or "tends to" instead of "converge towards", at least the singular "converges" should be used.

The wording has been changed to 'approaches the "true" profiles' as suggested (Page 10, line 12).

Page 11, line 2–3: I'd suggest to remove "are" at the beginning of line 3 to read "The lowest AVK peaks at ..."

The suggested change has been made (Page 11, lines 19–20).

Page 11, line 3: I'd suggest to change to ". . . where the MR is larger than ... ".

The suggested change has been made (Page 11, line 20).

Page 11, line 5: I'd suggest to insert "the" in front of "OH".

The suggested change has been made (Page 11, line 22).

Page 11, lines 6–7: As noted above in the ozone section, a resolution that changes to larger numbers means that it gets worse and is therefore decreasing. Secondly, there is not much lower thermosphere left below 90 km, so that expression should probably removed here.

The suggested changes have been made as follows (Page 11, lines 24–25)

"… altitude resolutions decreasing from 10.2 km in the lower mesosphere to 16.6 km in the upper mesosphere, at altitudes below 90 km."

Page 11, line 10: I'd suggest to change "small" to "smaller".

The suggested change has been made (Page 11, line 27).

Page 11, line 13: I'd suggest to insert "the" before "MC".

The suggested change has been made (Page 11, line 31).

Page 11, line 17: "are dependent" can be shortened to "depend".

The suggested change has been made (Page 12, line 3).

Page 11, lines 26–28: This sentence is unclear and confusing, should "to" be "as"? It is confusing which signal changes and how OH relates to ozone, and how that relates to the receiver performance. I'd suggest to not put everything into a single sentence.

The text has been re-written as follows to make it clearer (Page 12, lines 11–15):-

"The predicted changes in OH VMR, by up to ~20 ppbv for the altitude range 64–84 km, will have a small effect on the measured atmospheric brightness temperatures, producing a ~10 mK increase close to the emission line centre. However, the enhanced OH microwave signal during sub-storm activity would be measurable with a baseline noise of 1 mK for a 6 hour integration."

Page 12, line 3: I'd suggest to remove "seasons".

The suggested change has been made (Page 12, line 21).

Page 12, line 4: I'd suggest to change "differing" to "different".

The suggested change has been made (Page 12, line 22).

Page 12, line 5: I'd suggest to use "performing" instead of "making" and to insert "a" (or "an") in front of "rms".

The suggested changes have been made (Page 12, lines 23–24).

Page 12, line 6: According to the main text, the ozone altitude resolution is closer to 11–18 km.

Thank you for spotting this mistake. The range of ozone altitude resolutions has been changed to the correct values, 11–18 km (Page 12, line 24).

Page 12, line 18: I'd suggest to remove the commas around "and man-made".

The suggested change has been made (Page 13, line 29).

Page 12, line 19: I'd suggest to change "making observations" to "observing" or "performing observations".

The wording has been changed to 'observing' as suggested (Page 13, line 30).

Page 12, line 19: I believe "or polar" is not needed and can be removed, "high latitude" already includes "polar".

The suggested change has been made (Page 13, line 31).

Page 12, line 20: I'd suggest to change to: ". . . and by carefully pointing the receiver ... ".

The suggested change has been made (Page 13, line 31).

Page 12, line 21–22: I'd suggest to remove "values of the" and to insert "need to be" before "verified".

The suggested changes have been made (Page 13, line 33).

Page 12, line 23: I'd suggest to change "that" to "which".

The suggested change has been made (Page 13, line 34).

Page 12, line 24: I'd suggest to change "appropriate" to something like "more appropriate" or "a better approach" or something similar.

The wording has been changed to "a better approach may be …" (Page 14, line 1).

Page 12, line 28 (data availability): I'd suggest to be more specific: "The model and simulation datasets used in this study ... ".

The suggested change has been made (Page 14, line 5).

Figures

Fig. 6: It would be helpful to put titles to the panels themselves, "sub-storm" on the left and "background" on the right. The authors may consider swapping the order since "background" has the connotation of being the usual state, i.e. the normal state when nothing happens.

The suggested changes to the figure have been made as shown below (Fig. 6).

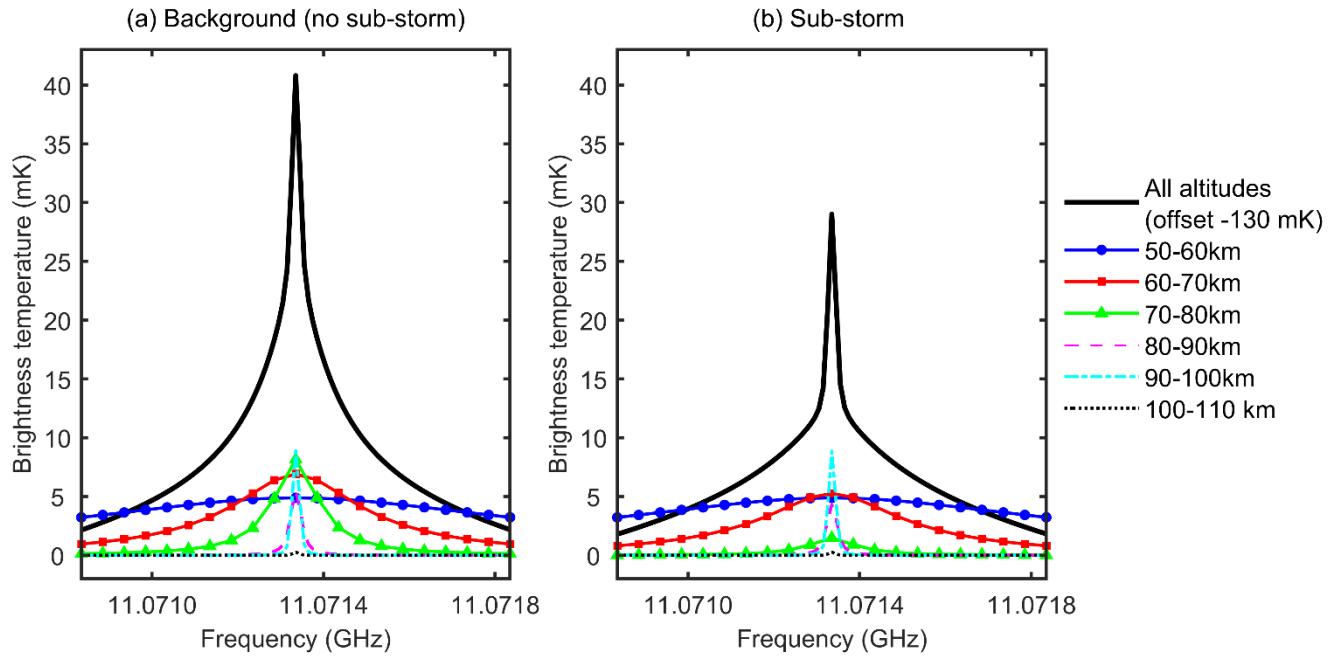



Fig. 7: I'd suggest to label the panels, i.e. the columns and rows such that is much easier to the cases without reading the rather long figure caption. Similar to Fig. 6 that would mean indicating the columns with "sub-storm" and "background" (consider swapping, see above), and the rows with "both", "vertical", and "horizontal".

The suggested changes to the figure have been made as shown below (Fig. 7).

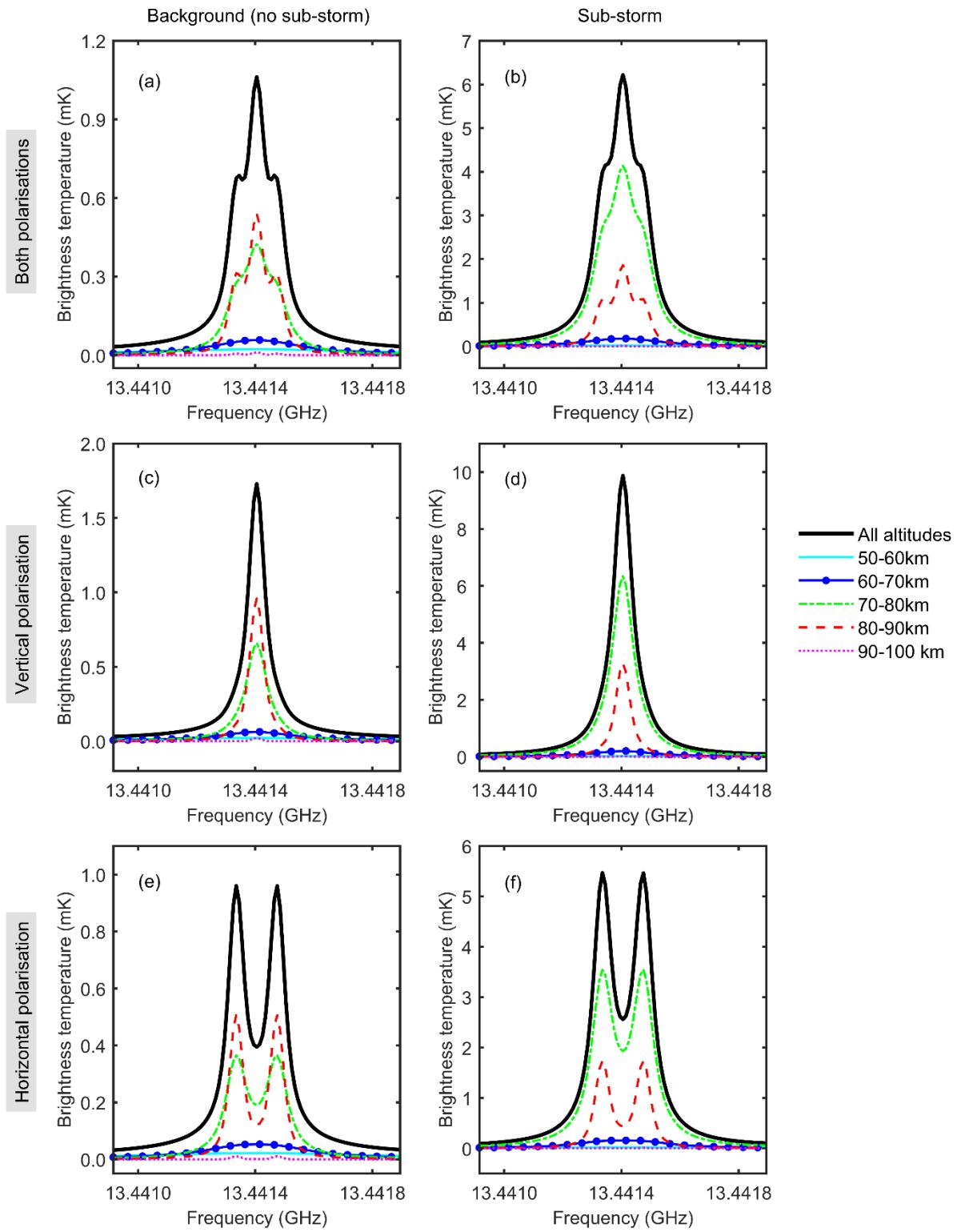



Fig. 9, caption, lines 5–6: I believe that the commas after "kernels" and before "are shown" should be removed.

The suggested change has been made (Fig. 9, caption, lines 5–6).

Fig. 10: The light yellow line is hard to see in prints, please use a different colour.

The suggested change to the figure has been made as shown below (Fig. 10). The colours of some of the other lines have also been changed.

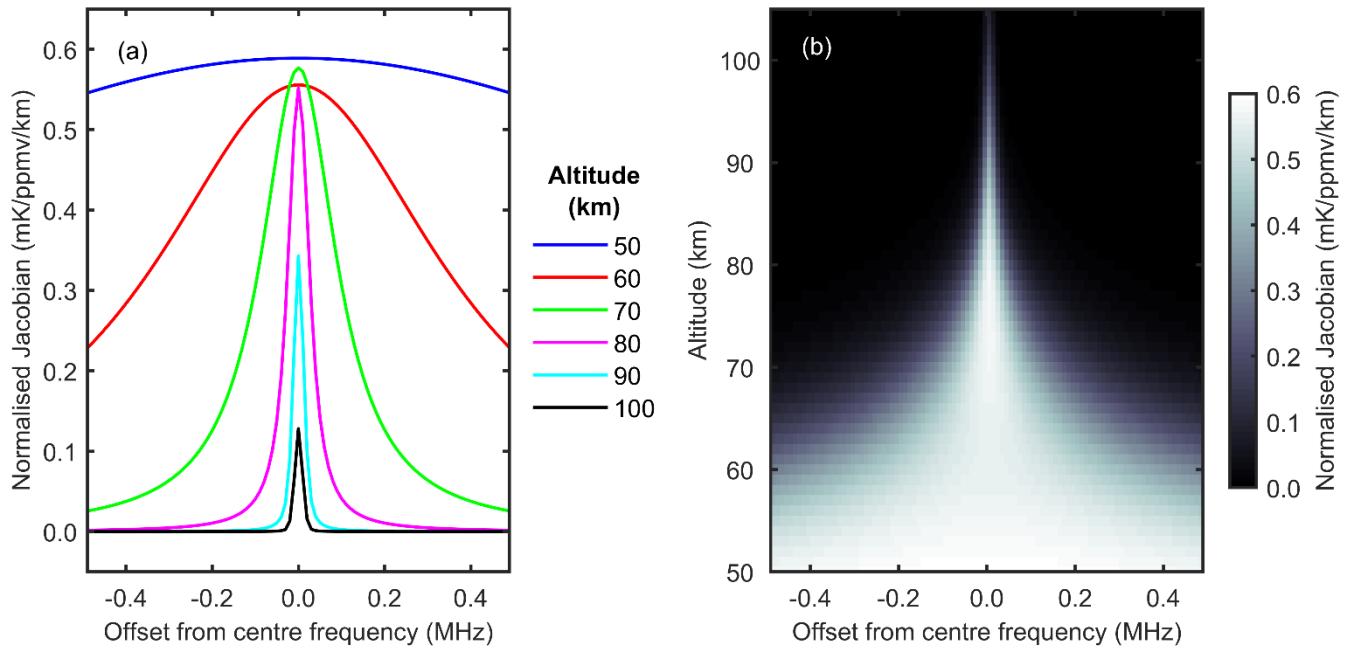
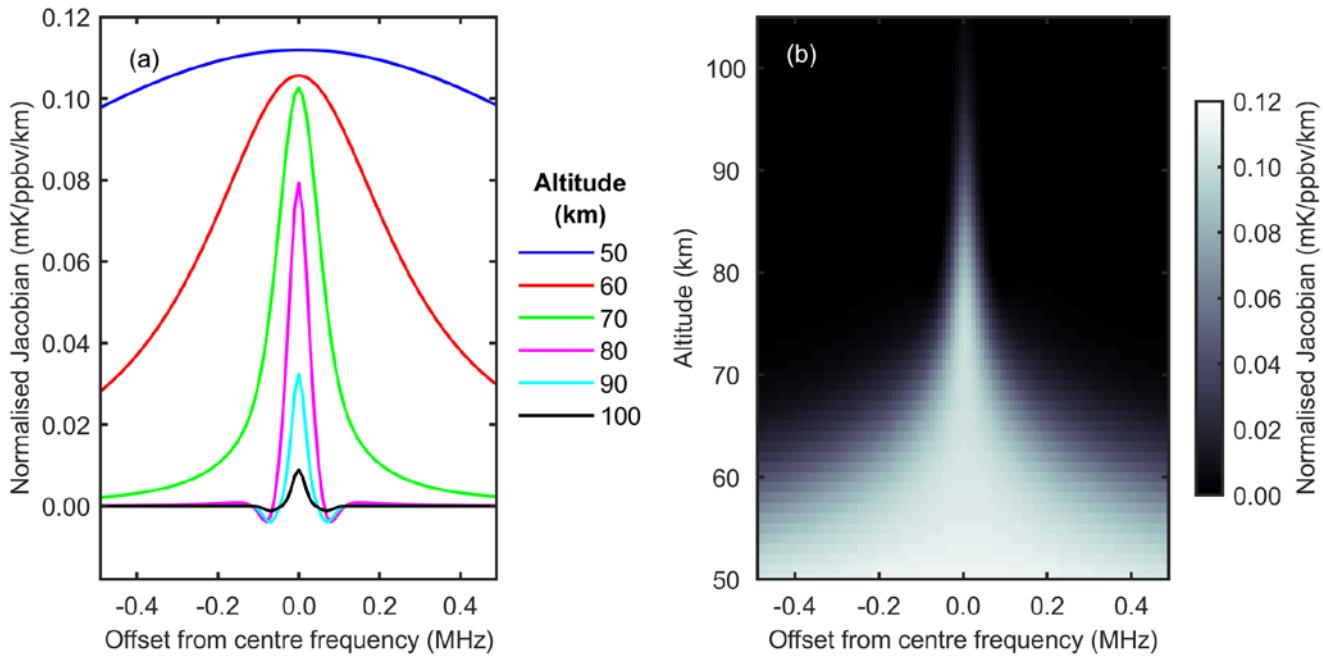



Fig. 12, caption, line 6: I believe that the commas after "kernels" and before "are shown" should be removed.

The suggested change has been made (Fig. 12, caption, line 6).

Fig. 13: The light yellow line is hard to see in prints, please use a different colour.

The suggested change to the figure has been made as shown below (Fig. 13). The colours of some of the other lines have also been changed.

Other changes to the manuscript

An additional co-author, Annika Seppälä, has been added to the author list (Page 1, lines 3–8). The Acknowledgments section has been revised accordingly (Page 14, lines 12–13). The journal editor and *Copernicus* editorial office have been notified and they have confirmed that this change can be made.

The reference "Rodger, C. J., et al., J. Geophys. Res., 120, doi:10.1002/2015JA021537, 2015." is not needed and has been removed (Page 16, line 32).

Other changes have been made in response to comments by Anonymous Referee #2 and Anonymous Referee #3.

Simulation study for ground-based Ku-band microwave observations of ozone and hydroxyl in the polar middle atmosphere

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10 **Abstract.** The Ku-band microwave frequencies (10.70–14.25 GHz) overlap emissions from ozone (O_3) at 11.072 GHz and hydroxyl radical (OH) at 13.441 GHz. These important chemical species in the polar middle atmosphere respond strongly to high-latitude geomagnetic activity associated with space weather. Atmospheric model calculations predict that energetic electron precipitation (EEP) driven by magnetospheric sub-storms produces large changes in polar mesospheric O_3 and OH. The EEP typically peaks at geomagnetic latitudes ~65° and evolves rapidly with time longitudinally and over the geomagnetic
15 latitude range 60°–80°. Previous atmospheric modelling studies have shown that during sub-storms OH abundance can increase by more than an order of magnitude at 64–84 km and mesospheric O_3 losses can exceed 50%. In this work, an atmospheric simulation and retrieval study has been performed to determine the requirements for passive microwave radiometers capable of measuring diurnal variations in O_3 and OH profiles from high-latitude northern hemisphere and Antarctic locations to verify model predictions. We show that, for a 11.072 GHz radiometer making 6 h spectral measurements
20 with 10 kHz frequency resolution and root-mean-square baseline noise of 1 mK, O_3 could be profiled over 8×10^{-4}–0.22 hPa (~98–58 km) with 10–17 km height resolution and ~1 ppmv uncertainty. For the equivalent 13.441 GHz measurements with vertical sensor polarisation, OH could be profiled over 3×10^{-3}–0.29 hPa (~90–56 km) with 10–17 km height resolution and ~3 ppbv uncertainty. The proposed observations would be highly applicable to studies of EEP, atmospheric dynamics, planetary-scale circulation, chemical transport, and the representation of these processes in polar and global climate models.
25 Such observations would provide a relatively low-cost alternative to increasingly sparse satellite measurements of the polar middle atmosphere, extending long-term data records and also providing "ground truth" calibration data.

1 Introduction

1.1 Background information

Energetic particle precipitation (EPP) is an important mechanism in the polar middle and upper atmosphere, causing ionisation
30 in the neutral atmosphere and producing odd nitrogen ($NO_x = NO + NO_2$) and odd hydrogen ($HO_x = OH + HO_2$) (Brasseur

and Solomon, 2005; Mironova et al., 2015; Sinnhuber et al., 2012; Verronen and Lehmann, 2013). Enhanced abundances of these chemical species lead to catalytic destruction of ozone (O_3) (Jackman and McPeters, 2004), perturbing the radiative balance, dynamics, and large scale circulation patterns of the atmosphere. This mechanism potentially links solar variability associated with space weather to regional surface climate (e.g. Arsenovic et al., 2016; Baumgartner et al., 2011; Semeniuk et

5 al., 2011; Seppälä et al., 2009, 2013). The energetic particles, mainly protons and electrons of solar and magnetospheric origin, vary widely in energy range and the regions of the atmosphere where they impact, both in geographic/geomagnetic coverage and altitude. Energetic electron precipitation (EEP), with electron energies in the range 20–300 keV, increases ionization in the polar mesosphere at altitudes of 60–90 km (Newnham et al., 2018; Turunen et al., 2009).

Atmospheric model calculations (Seppälä et al., 2015) predict that EEP driven by magnetospheric sub-storms produces large

10 changes in polar mesospheric O_3 and HO_x. The EEP typically peaks at geomagnetic latitudes ~65° (e.g. Kilpisjärvi, Finland and Syowa station, Antarctica) and evolves rapidly with time eastwards and over the geomagnetic latitude range 60°–80° (Cresswell-Moorcock et al., 2013). During the sub-storms the modelled night-time OH partial column over the altitude range 64–84 km OH volume mixing ratio (VMR) can increase by more than 1000% at 64–84 km and the OH volume mixing ratio (VMR) at 70 km increases from the background (i.e. no sub-storm) level of ~8 ppbv to ~60 ppbv (Seppälä et al., 2015). The

15 sub-storms leave footprints of 5–55% mesospheric O_3 loss lasting many hours of local time, with strong altitude and seasonal dependences (Seppälä et al., 2015). The cumulative atmospheric response of ~1250 sub-storms.yr^{-1} (Rodger et al., 2016) is potentially more important than the impulsive but highly sporadic (~3–4/year) effects of solar proton events. Other recent studies suggest that EEP from the Earth's outer radiation belt continuously affects the composition of the polar mesosphere through HO_x-driven chemistry (Andersson et al., 2014a), and that HO_x may cause prolonged mesospheric O_3 depletion when

20 NO_x is enhanced during polar winter (Verronen and Lehmann, 2015). Mesospheric OH is predominantly produced by the photo-dissociation of water vapour (H_2O) and HO_x measurements can also be used as a proxy for mesospheric H_2O (Summers et al., 2001). In order to test, verify, and improve models of the polar stratosphere and mesosphere, O_3 and OH observations are needed with sufficient precision and time resolution to characterise the different processes that modify their chemical abundances. This paper identifies the instrument characteristics that would be needed to produce the required observations.

25 **1.2 Previous mesospheric ozone and hydroxyl measurements**

O_3 vertical profiles retrieved in the upper mesosphere (70–100 km) from observations by nine recently-operating satellite instruments have been reviewed and compared (Smith et al., 2013). The comparison of coincident profiles showed that upper mesospheric ozone is much more abundant during the night than and depleted during the day, the secondary O_3 VMR maximum occurs at 90–92 km during the day and 95 km at night, and O_3 VMR is very low (<0.2 ppmv) at about 80 km during

30 both day and night with a minimum in ozone density at sunrise at 80 km. Another instrument, tThe Microwave Limb Sounder (MLS) on the Aura satellite, has provided a long time series of stratospheric and mesospheric O_3 measurements (Froidevaux et al., 2008) with global coverage, although the precision of retrieved O_3 profiles decreases sharply above 0.1 hPa (~64 km). MLS observations of mesospheric OH (Pickett et al., 2008) have also helped to elucidate the role of different types of EPP in

polar O_3 variability (e.g., Andersson et al., 2014b; Verronen et al., 2011; Zawedde et al., 2018). Minschwaner et al. (2011) reviewed OH in the stratosphere and mesosphere and used MLS data to study its diurnal variability. The Spatial Heterodyne Imager for Mesospheric Radicals (SHIMMER) (Englert et al., 2010) measured OH diurnal variations for investigations into discrepancies in mesospheric HO_x in photochemical models that had been suggested by earlier satellite observations (Siskind et al., 2013). A major limitation of satellite observations is that the temporal and spatial sampling arising from several overpasses per day at polar locations can make investigating rapidly-evolving, short term, chemical changes, such as those induced by sub-storms, a challenge.

Ground-based millimetre-wave radiometry at 110–250 GHz provides continuous measurements of O_3 (e.g., Hartogh et al., 2004; Daae et al., 2014; Ryan et al., 2016) and perhydroxyl radical (HO_2) (Clancy et al., 1994) but the altitude range is typically restricted to ~20–75 km. Above ~75 km, thermal Doppler broadening increases and lower pressure / higher altitude information cannot be retrieved from the emission lines. However, for the O_3 microwave line centred at 11.072 GHz the atmosphere is much less opaque and Doppler broadening is 10–23 times lower than at 110–250 GHz, allowing the retrieval of O_3 VMR to higher altitudes. Low cost, ground-based microwave radiometers operating at 11.072 GHz have been developed using inexpensive Ku-band satellite television low noise block (LNB) downconverters (Rogers et al., 2009; Rogers et al., 2012). Applying a straightforward processing scheme, O_3 partial columns for the lower mesosphere (~50–80 km) and the upper mesosphere / lower thermosphere (~80–100 km) have been determined from the central 1.25 MHz section of observed 11.072 GHz O_3 spectra. The current observations can broadly estimate seasonal O_3 variability near the mesopause but would not readily resolve the altitude-dependent O_3 changes at 60–90 km that are predicted to occur with sub-storms. Furthermore, due to the weakness of the 11.072 GHz emission line, measurement times extend to days using a single receiver radiometer achieving a root mean square (rms) noise level (1σ) of 5 mK at 9.8 kHz resolution and with 24 h signal integration.

Remote sensing measurements of mesospheric OH abundances from the ground are challenging due to the low VMR, which typically peaks in the ppbv range at ~80 km. Ground-based LIDARs at mid-latitude sites have detected and identified OH in the ground vibrational state, $X^2\Pi$ (v" = 0), in the mesosphere at ~75–85 km altitude (Brinksma et al., 1998). The LIDAR measurements detect UV resonant fluorescence at ~308 nm from the OH $A^2\Sigma – X^2\Pi$ electronic transition. In contrast, near-infrared and visible airglow emissions in the $X^2\Pi$ ro-vibrational Meinel system are due to deactivation of vibrationally-excited OH* (v' = 7 to v' = 9) generated by the highly exothermic reaction of atomic hydrogen with O_3. Satellite measurements of Meinel band night-glow have shown that OH* occurs in a ~8 km thick layer near 90 km (Zhang and Shepherd, 1999). Ground-based spectroscopic measurements of individual OH vibrational bands in the Meinel emissions can be used to infer temperatures at the mesopause region and estimate relative populations of the v' states (e.g., von Zahn et al., 1987; Yee et al., 1997; Smith et al., 2010).

1.3 This work

In this work we investigate the potential for measuring diurnal variations in the vertical profiles of O_3 in the mesosphere and lower thermosphere using the 11.072 GHz emission line, and extending the ground-based microwave radiometry technique to

mesospheric OH emissions at ~13.4 GHz. The microwave spectrum of the most abundant hydroxyl isotopomer, ^{16}OH, at 13.433–13.442 GHz shows four closely-spaced lines which arise from the Λ-doubling hyperfine structure in the rotational $^2\Pi_{3/2}$ (v = 0, J = 7/2) state (Radford, 1961; Sastry and Vanderlinde, 1980). The atmospheric OH spectrum is further complicated by the magnetic Zeeman effect, due to the molecule's non-zero total electron spin quantum number (S = 1/2).

5 Each line is Zeeman split into several components polarised in a quasi-symmetric manner and shifted from the central frequency. Here we investigate potential measurements using the most intense of the four OH rotational lines, which has a line position of 13.441 GHz.

The study focuses on atmospheric simulations and retrievals for Kilpisjärvi, Finland, since model data for O_3 and OH abundances in sub-storm and background (no sub-storm) conditions are available for this location. However, we demonstrate

10 the technique's wider applicability by comparing atmospheric microwave transmittances at six land-based locations including Kilpisjärvi, shown on the maps in Figure 1. Three potential sites for ground-based instruments are in the Antarctic and three are at high-latitude in the northern hemisphere (NH), and all are located close to geomagnetic latitude 65° where sub-storms are predicted to have the greatest effect on mesospheric O_3 and OH.

2 Methodology

15 Microwave spectrum simulations and retrievals have been performed using atmospheric model data-sets in radiative transfer calculations for selected high-latitude and polar locations. The synthesis of VMR profiles at these locations for O_3, OH, and seven other atmospheric species, as well as temperature profiles, from available model data is described in Section 2.1. The configuration of the radiative transfer forward model for simulating clear-sky atmospheric microwave transmittance and brightness temperature spectra is given in Section 2.2, and the setups for performing O_3 and OH retrievals for Kilpisjärvi,

20 Finland are given in Section 2.3.

2.1 Atmospheric model datasets

Nine chemical species were identified as significant contributors to the clear-sky atmospheric microwave spectrum in the 11–14 GHz region, overlapping the target O_3 and OH lines at 11.072 GHz and 13.441 GHz: ozone (O_3), hydroxyl radical (OH), water vapour (H_2O), molecular nitrogen (N_2), molecular oxygen (O_2), perhydroxyl radical (HO_2), nitric acid (HNO_3), hydrogen

25 peroxide (H_2O_2), and carbon dioxide (CO_2). Monthly mean vertical profiles of VMR for the atmospheric species, and temperature, were calculated using a 10-year dataset from WACCM-D (Verronen et al., 2016) covering 2000–2009 from WACCM-D (Verronen et al., 2016). WACCM-D is a 3-D global atmospheric model that incorporates a detailed representation of *D*-region chemistry in the specified dynamics (SD) version of the Whole Atmosphere Community Climate Model (WACCM 4) (Marsh et al., 2013). Model The model data were at taken from the WACCM-D longitude / latitude grid points closest to

30 the six locations of interest. Seasonal winter mean profiles were calculated using December, January, and February (DJF) data

for the NH locations and June, July, and August (JJA) data for the southern hemisphere (SH). Similarly, summer mean profiles were calculated using JJA data for the NH locations and DJF data for the SH locations.

O_3 and OH VMR profiles for sub-storm and background (no sub-storm) conditions were computed by combining data from the 1-D Sodankylä Ion and Neutral Chemistry (SIC) model (Verronen et al., 2005) with monthly-mean WACCM-D data. The

5 SIC model data provided VMR data over the altitude range 20–100 km and monthly-mean data from WACCM-D filled the region from the ground up to 20 km and above 100 km. Figure 2(a) shows O_3 number density profiles calculated using a 9-day SIC model run (Seppälä et al., 2015) for December 2007 with sub-storm conditions at Kilpisjärvi. The largest decreases in mesospheric O_3 abundance, exceeding 50%, occur over the altitude range 68–86 km during a four-day period ~~over the altitude range 68–86 km~~, after which the number densities return to background levels (Figure 2(b)). The percentage changes in O_3

10 partial column over altitudes 64–84 km for the original (15 min) model resolution and 1 h, 6 h, and 24 h moving average smoothed data are shown in Figure 2(c). The three smoothed datasets show reduced diurnal variability compared to the original model data but the largest decrease in O_3 partial column is little changed by ~~a moving~~ averag~~e of~~ing up to 24 h. VMR profiles for sub-storm and background (no sub-storm) conditions were determined from the SIC model data coincident with the largest decrease in O_3 partial column. ~~For the 6 h smoothed SIC data the largest decrease in O_3 partial column occurred between~~

15 ~~01:30–07:30 UTC on day 363 (29 December). Sub-storm, background (no sub-storm), and~~, and these and the monthly-mean ~~O_3 VMR~~ profiles are shown in Figure 2(d). Sub-storm reductions in O_3 VMR occur ~~around~~ over the altitude range 64–94 km, with the largest decreases at ~~75~~ 72 km. ~~Compared to the monthly-mean data, O_3 VMR from the SIC model runs is ~2× higher in the stratosphere above 20 km and for the secondary ozone layer in the upper mesosphere at ~95 km for this particular scenario.~~

20 A similar approach was used to determine OH VMR profiles, as shown in Figure 3. The largest increases in mesospheric OH abundance, exceeding two orders of magnitude above the background level, occur at altitudes in the range 66–82 km during the four days corresponding to O_3 decreases, after which OH number densities return to background levels (Figures 3(a) and 3(b)). The percentage changes in OH partial column over altitudes 64–84 km for the original (15 min) model resolution and 1 h, 6 h, and 24 h moving averages are shown in Figure 3(c). The enhanced level of mesospheric OH during sub-storms shows

25 greater diurnal variability than the corresponding O_3 reductions. Smoothing the modelled OH partial columns significantly reduces the daily maxima, indicating that the improved signal-to-noise from atmospheric measurements longer than ~6 h would be strongly offset by the smaller integrated OH signal. ~~For the 6 h smoothed SIC data the largest decrease in OH partial column occurred between 04:45–10:45 UTC on day 362 (28 December).~~ The ~~corresponding~~ 6-h smoothed sub-storm and background (no sub-storm) VMR profiles corresponding to the largest decrease in OH partial column,~~,~~ and monthly-mean~~,~~

30 ~~OH VMR~~ profiles, are shown in Figure 3(d). The sub-storm profile shows a five-fold increase in OH VMR around 82 km compared to the monthly-mean and background (no sub-storm) data.

2.2 Forward modelling of atmospheric spectra

Simulated atmospheric microwave spectra were calculated using version 2.2.58 of the Atmospheric Radiative Transfer Simulator (ARTS) (available at http://www.radiativetransfer.org/) (Buehler et al., 2005; Buehler et al., 2018; Eriksson et al., 2011) and the Qpack2 (a part of atmlab v2.2.0) software package (Eriksson et al., 2005). ARTS is a monochromatic line-by-line model that can simulate radiances from the infrared to the microwave and has been validated against other models in the submillimetre spectral range (Melsheimer et al., 2005). The model includes contributions from spectral lines and broadband continua via a choice of user-specified parameterisations. The Planck formalism was used for calculating brightness temperatures and atmospheric transmittance.

Spectroscopic line parameters for O_3, OH, H_2O, N_2, O_2, HO_2, HNO_3, H_2O_2, and CO_2 were taken from the high-resolution transmission (HITRAN) molecular absorption database (Gordon et al., 2017). For all molecules except OH the Kuntz approximation (Kuntz, 1997) to the Voigt lineshape was used with a Van Vleck–Huber prefactor (Van Vleck and Huber, 1977) and a line cut-off of 750 GHz which is valid for the pressures considered. ~~The~~ As the water vapour continuum parameterisation ~~used was~~ the Mlawer–Tobin Clough–Kneizys–Davies (MT-CKD) model (version 2.5.2) was used, which ~~separately~~ includes both foreign and self-broadening components (Mlawer et al., 2012). Collision-induced absorption (CIA) is the main contribution to the dry continua in the microwave range, and ~~hence~~ therefore the CIA parameterisations from the MT-CKD model (Clough et al., 2005) (version 2.5.2 for N_2 and CO_2 and version 1.0 for O_2) were applied.

In order to compare seasonal atmospheric transmittances at the six different locations, survey spectra over the frequency range 5–20 GHz were calculated at zenith angles of 0° and 82° using mean winter and mean summer profiles. A 1 MHz frequency grid was chosen, adequate to characterise smoothly varying, broadband transmittance but insufficient to resolve narrow-band spectral features.

For simulations of ground-based microwave measurements, values for the frequency resolution, bandwidth, and baseline noise were chosen by considering the range of atmospheric emission linewidths and likely instrument performance. Pressure-broadened, Doppler-broadened, and Voigt full-width half-maxima (FWHM) linewidths for the lines were calculated using air-broadening coefficients from HITRAN and winter (DJF) and summer (JJA) pressure and temperature profiles at Kilpisjärvi. Figures 4(a) and 4(b) show the variation of O_3 11.072 GHz and OH 13.441 GHz linewidths with altitude. For both molecules pressure-broadening dominates below ~90 km, increasing rapidly below 85 km to give FWHM linewidths of 300 kHz at ~65–70 km in winter and ~70–75 km in summer. Voigt linewidths are lowest in the lower thermosphere at ~95–100 km, with minimum FWHM of 17 kHz for O_3 and 34 kHz for OH, and increase above 100 km due to increased Doppler broadening. Forward model spectra were therefore calculated with a frequency grid spacing of 10 kHz over a bandwidth of 1 MHz to encompass the range of emission linewidths from O_3 and OH in the mesosphere and lower thermosphere. All measurement simulations were performed for a single pencil beam of radiation at a zenith angle of 82°.

An important consideration in our calculations is that O_3 and OH atmospheric spectra can be measured over 6 h with rms baseline noise of 1.0 mK, and this assumption is justified as follows. Low-cost LNB receivers with noise factor ~1.3 dB

currently used in ~~MOSAIC~~ O$_3$ 11.072 GHz spectrometers for the educational project Mesospheric Ozone System for Atmospheric Investigations in the Classroom (MOSAIC) operate uncooled, i.e. at ambient temperature (Rogers et al., 2012). By analysing signals from both horizontal and vertical polarisation channels of the LNB with 10 kHz frequency resolution, and integrating the atmospheric signal for 24 h, an atmospheric spectrum with baseline noise of ~3.4 mK is achieved. The

5   statistical fluctuation ΔT (K) in the total system temperature, T_{sys} (K), is calculated according to the ideal radiometer equation (Kraus, 1986):

$$\Delta T = \frac{T_{sys}}{\sqrt{t \Delta f N_{CH}}} \qquad\qquad\qquad (1)$$

10   where t is observation time (in s), Δf is the frequency resolution (in Hz) of the radiometer, and N_{CH} is the number of measurement channels. Applying this equation we can define the sensor characteristics needed to achieve a signal-to-noise of 1.0 mK in 6 h measurements, providing adequate time resolution to observe predicted changes in mesospheric O$_3$ and OH. The results in Table 1 show that higher performance LNB receivers, with noise factor 0.7 dB, would achieve such measurements of the O$_3$ 11.072 GHz emission if cooled to 168 K. Alternatively an array of 12 receivers operating at room

15   temperature, with a total of 24 measurement channels, would achieve this measurement performance. At the higher frequency (13.441 GHz) of the OH emission available LNBs are less sensitive, with noise factor 1.2 dB, and would require cryogenic operation at 46 K to achieve the equivalent performance. A receiver array at room temperature would need 40 spectrometers (i.e. 80 measurement channels) to achieve the required measurement performance at 13.441 GHz. Thus we conclude that suitable receivers could be constructed using commercially-available LNB receivers, albeit with the added complexity and

20   higher power consumption needed for low-temperature operation or, alternatively, by combining the outputs from multiple room temperature receivers to achieve higher signal-to-noise spectra.

The treatment of the Zeeman effect in the Stokes formalism of ARTS is described by Larsson et al. (2014). The Zeeman components of the OH line centred at 13.441 GHz were calculated for an instrument viewing at an azimuthal angle 0° i.e., northward-looking from Kilpisjärvi, with sensor polarisation either vertical, horizontal, or in both directions combined.

25   Magnetic field data were taken from IGRF-11 (Finlay et al., 2010) and the ~~renormalised~~ Faddeeva function was used to describe the lineshape.

Forward model spectra were calculated for ground-based, clear-sky observations during December sub-storm and background (no sub-storm) conditions at Kilpisjärvi. The emission signals from different altitudes were found by selectively setting O$_3$ and OH VMR to zero for 10 km sections of the mesosphere and lower thermosphere, and also running simulations with zero

30   O$_3$ and OH VMR at all altitudes. In order to assess measurement uncertainties in the retrieval algorithm and its ability to reproduce the "true" state of the atmosphere, two sets of 500 spectra each of O$_3$ and OH were calculated for Monte Carlo (MC) error analysis. In one set of MC repeat spectra all VMR profiles were kept constant at the "true" values whereas in the other

set the O_3, OH, and H_2O profiles were randomly scaled ~~by~~ using a uniform distribution over the range 0.5–2.0. Baseline noise with rms level 1.0 mK was randomly calculated and added to each individual O_3 and OH spectrum in both of the MC sets.

2.3 Retrievals

5 The 6 h atmospheric spectra, simulated for December sub-storm and background (no sub-storm) conditions at Kilpisjärvi, were inverted into altitude profiles of O_3 and OH VMR using the optimal estimation method (OEM) (Rodgers, 2000) implemented in Qpack (Eriksson et al., 2005). Iterative absorption calculations in ARTS were performed line-by-line ~~inside~~ within the radiative transfer calculation, rather than using pre-calculated look-up tables, in order to accurately model atmospheric spectra (Buehler et al., 2011). VMR values were retrieved ~~over~~ for altitude levels 0–120 km with a 1 km spacing, where hydrostatic equilibrium ~~is~~ was assumed for the altitude and pressure. The O_3, OH, and H_2O a priori VMR profiles were the December 10 monthly mean profiles for Kilpisjärvi. The diagonal elements in the covariance of the O_3 and OH a priori were fixed ~~at~~ to 1.5 ppmv and 10 ppbv respectively whereas for H_2O they were fixed at the square of 50% of the VMR values. The off-diagonal elements ~~The shapes~~ of the covariance ~~were set to~~ linearly decrease ~~towards the off-diagonal elements~~ with a correlation length of a fifth of a pressure decade (approximately 3 km).

3 Results

15 ### 3.1 Simulated atmospheric spectra

Survey clear-sky atmospheric transmittance spectra, calculated for the selected locations, are shown in Figure 5. Water vapour absorption leads to decreased transmittance at frequencies above 10 GHz. Higher transmittance occurs for the three colder, desiccated Antarctic sites whereas the generally milder NH locations have lower transmittance especially in summertime. Considering the frequencies of the O_3 and OH lines, at zenith angle 0°, transmittance is 0.98 at the six locations in both winter 20 and summer conditions. At a zenith angle 82° the lower transmittance, in the range 0.84–0.86, is due to the higher air mass factor and increased tropospheric attenuation in particular by water vapour, oxygen, and nitrogen continua. However, the very similar transmittances at various locations suggest that the ~~differing~~ different seasonal meteorology should have little impact on the proposed O_3 and OH observations, unlike ground-based measurements using higher frequencies in the millimetre-wave region where cold, dry, high-altitude sites are advantageous.

25 Forward model simulations of the O_3 11.072 GHz emission line are shown in Figures 6(a) and 6(b), for the sub-storm and background (no sub-storm) cases respectively. The solid black lines in each plot are the brightness temperature spectra that would be observed at the ground in clear-sky conditions at a zenith angle of 82°. The other lines show the contributions to the total signal from 10 km layers in the mesosphere and lower thermosphere between 50 km and 110 km. For the sub-storm scenario, O_3 emission is reduced around the peak at 11.072 GHz compared to the background case. The largest decrease in

signal corresponds to altitudes 70–80 km, where the modelled reduction in O_3 number density during sub-storms is greatest, with smaller decreases at 60–70 km and 80–90 km.

Forward model simulations of the OH 13.441 GHz emission line are shown in Figures 7(a)–(f). Figures 7(a), 7(c), and 7(e) are the sub-storm spectra for three different sensor polarisations: - both polarisations, vertical, and horizontal respectively.

Figures 7(b), 7(d), and 7(f) show the corresponding spectra for the background (no sub-storm) case. The solid black lines in each plot are the brightness temperature spectra that would be observed at the ground in clear-sky conditions at a zenith angle of 82°. The other lines are the contribution to the total signal from 10 km layers in the mesosphere and lower thermosphere between 50 km and 100 km. At azimuthal angle 0° the vertical polarisation signal is dominated by a single line centred at 13.441 GHz whereas in horizontal polarisation the pair of Zeeman split lines are clearly resolved. For the sub-storm scenario, OH emission increases around the peak positions compared to the background case. As for O_3, the largest changes in signal corresponds to altitudes 70–80 km, where the modelled increase in OH number density during sub-storms is greatest, with smaller increases at 80–90 km.

3.2 Ozone retrievals

Example retrieval results are shown in Figure 8 and Figure 9 for simulated clear-sky 6 h observations of the O_3 11.072 GHz line at 82° zenith angle from Kilpisjärvi in December sub-storm and background conditions using a radiometer providing 10 kHz frequency resolution. Figures 8a and 8e show the final retrieval fits, for the sub-storm and background cases respectively, which agree with the simulated atmospheric spectra to within the rms baseline noise level. Figures 8c and 8g compare the true and a priori O_3 VMR profiles with the retrieved profiles for the sub-storm and background cases respectively. The solid red lines and shaded red areas show the average and standard deviations of the retrieved profiles from 500 repeat MC runs with the O_3 and H_2O profiles used to calculate the spectra, shown by the dashed blue lines, kept constant at the "true" model values. For both the sub-storm and background cases the retrieved O_3 profiles converge close to the "true" profiles over the altitudes where information is obtained from the observations, shown by the thicker solid lines and shaded grey areas. The description of how the retrieval altitude range is determined is given in the next paragraph. The differences between the retrieved and "true" VMR profiles, shown in Figures 8d and 8h, are in the range -1.0 – 0.6 ppmv. Above and below the retrieval range the O_3 profiles tend towards the a priori values.

The averaging kernels (AVKs) for every sixth retrieved altitude are shown in Figure 9a. The AVKs describe the relationship between the true, a priori, and retrieved atmospheric states (Rodgers, ~~2004~~2000). None of the AVKs peak at pressure levels above 0.0013 hPa (~94 km) due to Doppler broadening dominating over pressure broadening. The lowest AVK peaks are at 0.18 hPa (~60 km). The sum of the AVKs at each altitude, called the measurement (or total) response (MR), represents the extent to which the measurement contributes to the retrieval solution as compared to the ~~amount of~~ influence of the a priori at that altitude (Christensen and Eriksson, 2013). The altitude range where the retrieved O_3 profile has a high degree of independence from the a priori is ~~estimated~~ identified by MR values higher than 0.8. The retrieval altitude range is 8×10^{-4}–

0.22 hPa (~98–58 km), shown by the thicker sections of the lines and the shaded grey areas in Figure 8 and Figure 9. Outside of these altitudes (i.e. below 58 km and above 98 km) the MR weakens and the O_3 values in these regions should be interpreted with caution as the information from the a priori becomes important. The AVKs indicate the range of altitudes over which ~~the retrieved~~ O_3 ~~has smoothed the information in the data~~is observed. In the ideal case the AVKs would be delta functions but in

5   practice they are peaked functions with finite widths dependent on the spatial resolution of the observing system. ~~Thus, t~~The full-width half-maximum (FWHM) width~~s~~ of the kernels provide a measure of the vertical resolution of the retrieved profile. The FWHM values shown in Figure 9b indicate altitude resolutions ~~increasing~~ decreasing from 10.9 km in the lower mesosphere to 18.4 km in the upper mesosphere and lower thermosphere, at altitudes below 98 km. The altitude resolution can also be estimated from the degrees of freedom for signal (DOFS) for the inversion, given by the trace of the AVK matrix

10   (Rodgers, 2000; Ryan and Walker, 2015). Dividing the retrieved altitude range (~30 km) by the DOFS over the same range (~3.2) gives an altitude resolution of 12.0 km, similar to that obtained from the AVK FWHM values.

The OEM calculations provide observation errors (σ_{obs}) and total retrieval (observation plus smoothing) errors (σ_{tot}) ~~to give~~which provide further diagnostic uncertainty estimates of the ~~uncertainty of~~ retrieved profiles. The observation errors describe how the retrieved profiles are affected by measurement noise and are shown in Figure 9c, with typical values of about

15   0.34 ppmv. The observation errors are small outside of the range of the AVK peaks as the retrieval tends to the a priori values in these regions and the contribution from the measurement is small. The total retrieval errors shown in Figure 9d are in the range 1.09–1.34 ppmv, and ~~outside the range of AVK peaks~~ tend towards the smaller a priori standard deviations outside the range of AVK peaks.

The results of the MC error analysis using retrievals from 500 simulated spectra in which O_3 and H_2O profiles were randomly

20   scaled using a uniform distribution over the range 0.5–2.0~~were randomly scaled by 0.5–2.0~~, to test the retrieval algorithm's ability to reproduce the "true" state of the atmosphere, are shown in Figure 9e–f. Over the trustable altitude range the mean difference (μ_{MC}) between the MC retrieved and "true" profiles is in the range -0.8–2.0 ppmv. The standard deviation (σ_{MC}) of the individual retrievals is an estimator for the uncertainty of the O_3 retrieval, and the mean value of 0.8 ppmv is approximately double the mean observation error determined from single retrievals (σ_{obs}). Both parameters ~~are dependent~~depend on the

25   signal-to-noise ratio of the input spectrum but σ_{MC} may more realistically represent actual observations where mesospheric O_3 profiles and column H_2O show large variability.

The Jacobian and gain matrices ~~for of~~ the O_3 ~~retrievals~~forward model indicate ~~that the spectral response should be adequately sampled at~~ an instrument with 10 kHz frequency resolution ~~of 10 kHz~~will adequately sample the emission line for altitudes up to ~98 km where the O_3 measurement contributes significantly to the retrieval. The values of the Jacobian~~ describing the~~

30   ~~O_3 retrieval~~, normalised by the layer thickness of the retrieval grid for observations at an 82° zenith angle, are shown in Figure 10. Typical values of the Jacobian are ~0.5 mK (ppmv)$^{-1}$km^{-1} at mesospheric altitudes of 60–90 km where sub-storms are predicted to have the largest effect on O_3 abundance. The effect of predicted O_3 changes of ~1 ppmv over altitudes 68–86 km

on the measured atmospheric brightness temperature will therefore be small, of the order of 9 mK close to the emission line centre, but readily measurable with a baseline noise of 1 mK for a 6-hour integration.

3.3 Hydroxyl (OH) retrievals

Example retrieval results are shown in Figure 11 and Figure 12 for simulated clear-sky 6 h observations of the OH 13.441 GHz
5   line at 82° zenith angle and 0° azimuthal angle from Kilpisjärvi in December sub-storm conditions using a radiometer providing 10 kHz frequency resolution with vertical sensor polarisation. Similar results were obtained for retrievals with simulated spectra using sensors measuring horizontal or both polarisations, and with non-zero azimuthal angles. Figures 11a and 11b show the final retrieval fits which agree with the simulated atmospheric spectra to within the rms baseline noise level. Figures 11c and 11d compare the true sub-storm and a priori OH VMR profiles with the retrieved profiles. The solid red lines and
10   shaded red areas show the average and standard deviations of the retrieved profiles from 500 repeat MC runs with the OH and H_2O profiles used to calculate the spectra, shown by the dashed blue lines, kept constant at the "true" model values. The retrieved OH profile ~~converge towards~~approaches the "true" profiles over the altitudes where information is obtained from the observations, shown by the thicker solid lines and shaded grey areas. However, Figure 11d shows significant differences, in the range -13.6–3.3 ppbv, between the retrieved and "true" VMR profiles at altitudes which overlap the OH enhancements.
15   Convolving the true OH profile with the averaging kernels, to account for the limited retrieval resolution, results in much better agreement with differences (R-T*) in the range -1.4–2.9 ppbv. Above and below the retrieval range the OH profiles approach the a priori OH VMR values which are close to zero.

The averaging kernels (AVKs) for every sixth retrieved altitude are shown in Figure 12a. None of the AVKs peak at pressure levels above 0.003 hPa (~88 km) due to Doppler broadening dominating over pressure broadening. The lowest AVK peaks
20   ~~are~~at 0.24 hPa (~58 km). The estimated retrieval range, where the MR is ~~higher~~larger than 0.8, is 3×10^{-3}–0.29 hPa (~90–56 km), shown by the thicker sections of the lines and the shaded grey areas in Figure 11 and Figure 12. Outside of these altitudes (i.e. below 56 km and above 90 km) the MR weakens and the OH values in these regions should be interpreted with caution as the information from the a priori becomes important. The AVK FWHM values shown in Figure 12b indicate altitude resolutions ~~increasing~~decreasing from 10.2 km in the lower mesosphere to 16.6 km in the upper mesosphere ~~and lower~~
25   ~~thermosphere~~, at altitudes below 90 km. Dividing the retrieved altitude range (~34 km) by the DOFS over the same range (~3.0) gives an estimated altitude resolution of 11.0 km, similar to that obtained from the AVK FWHM values. The observation errors (σ_{obs}) shown in Figure 12c have typical values of 2.1 ppbv and are smaller outside of the range of the AVK peaks as the retrieval tends to the a priori values in these regions and the contribution from the measurement is small. The total retrieval errors (σ_{tot}) shown in Figure 12d are in the range 7.1–9.0 ppbv, and outside the range of AVK peaks tend towards
30   the larger a priori standard deviations.

The results of the MC error analysis using retrievals from 500 simulated spectra in which OH and H_2O profiles were randomly scaled by 0.5–2.0 are shown in Figure 12e–f. Over the trustable altitude range the mean difference (μ_{MC}) between the MC

retrieved and "true" profiles is in the range -1.4–1.6 ppbv. The standard deviation (σ_{MC}) of the individual retrievals is an estimator for the uncertainty of the OH retrieval, and the mean value of 2.6 ppbv is similar the mean observation error determined from single retrievals (σ_{obs}). Both parameters ~~are dependent~~depend on the signal-to-noise ratio of the input spectrum but σ_{MC} may more realistically represent actual observations where mesospheric OH profiles and column H_2O show

5   large variability.

The Jacobian and gain matrices ~~for~~ of the OH vertical polarisation ~~retrievals~~ forward model indicate that, as for O_3, th~~e~~ ~~spectral OH response~~emission line should be adequately sampled ~~at~~ by an instrument with 10 kHz frequency resolution ~~of 10 kHz~~ for all altitudes where the OH measurement contributes significantly to the retrieval. The values of the Jacobian ~~describing the OH retrieval~~, normalised by the layer thickness of the retrieval grid for observations at an 82° zenith angle, are shown in Figure

10   13. Typical values of the Jacobian are ~0.09 mK (ppbv)$^{-1}$km^{-1} at mesospheric altitudes of 60–90 km where sub-storms are predicted to have the largest effect on OH abundance. The ~~effect of~~ predicted changes in OH VMR~~changes,~~ by up to ~20 ppbv for the altitude range 64–84 km, will have a small effect~~,~~ on the measured atmospheric brightness temperature~~s is small~~, producing a ~~of the order of~~~-10 mK increase close to the emission line centre. ~~The~~However, the enhanced OH microwave signal during sub-storm activity ~~is of similar magnitude to the expected corresponding change in O_3 signal and, assuming~~

15   ~~similar receiver performance, should~~would be measurable with a baseline noise of 1 mK for a 6-hour integration.

4 Conclusions

The proof-of-concept simulations demonstrate that changes in O_3 and OH abundance in the high-latitude / polar middle and upper atmosphere, associated with geomagnetic sub-storm and other EEP processes, could be profiled using ground-based passive microwave measurements in the Ku-band 11–14 GHz region and OEM retrieval. At these frequencies tropospheric

20   attenuation is small and mesospheric emission signals are transmitted to the ground with low loss. Comparable, high atmospheric transmittances are calculated for various high-latitude NH and Antarctic sites in winter and summer~~seasons~~, suggesting that ~~differing~~different meteorology and location should have little impact on the proposed O_3 and OH observations. For a radiometer ~~making~~performing 6 h measurements of the 11.072 GHz O_3 emission line with 10 kHz frequency resolution and a rms baseline noise of 1 mK, O_3 could be profiled over 8×10^{-4}–0.22 hPa (~98–58 km) with ~~10–17~~11–18 km height

25   resolution (~12 km from DOFS analysis) and ~1 ppmv uncertainty. For the equivalent OH 13.441 GHz measurement, demonstrated for vertical sensor polarisation, OH could be profiled over 3×10^{-3}–0.29 hPa (~90–56 km), also with 10–17 km height resolution (~11 km from DOFS analysis) and ~3 ppbv uncertainty. Such observations would allow the diurnal variations in mesospheric O_3 and OH to be characterised when abundances are relatively high, e.g. during night-time and polar winter. Measurement times of 24 h or longer would be necessary with less sensitive receivers or for measuring lower

30   abundances. However, these measurements would still provide valuable data, in particular for the secondary ozone layer above 90 km as satellite measurements of this region become increasingly sparse. We have used simulations of atmospheric spectra

from Kilpisjärvi, Finland as a test case to assess the feasibility of such measurements but demonstrate that similar results would be achieved by instruments deployed at other high-latitude NH sites and in Antarctica.

LNB receivers operating at the Ku-band emission frequencies of O_3 and OH are commercially available-. LNBs covering the O_3 frequency (11.072 GHz) have noise factor typically in the range 0.7–1.3 dB, although suppliers specify values as low as

5   0.1 dB. For coverage of the OH emission line frequency (13.441 GHz) the stated LNB noise factor is ~1.2 dB. However, measurements reported by Tenneti and Rogers (2009) suggest that inexpensive satellite TV downconverters can achieve a noise factor as low as ~0.23 dB. If this high level of LNB performance is confirmed, a single receiver operating at room temperature would achieve 6 h O_3 spectrum measurements with the required rms baseline noise level of 1 mK. Otherwise, for the proposed O_3 and OH observations, either simultaneous measurements by an array of multiple receivers or using single

10 receivers cooled to ~168 K for O_3 and ~46 K for OH would be needed. Monitoring of O_3 in the same volume of the atmosphere using three LNB receivers mounted in front of a "Direct TV" offset parabolic dish has been reported (Rogers et al., 2012). With six spectrometers simultaneously measuring both polarisations from each of the three receivers, in the ideal case the improvement in signal-to-noise compared to a single radiometer is $\sqrt{6}$ = ~2.4, although non-optimum matching of the LNB beam profiles and antenna focus would impact on the performance. Scaling up to 24 independent spectrometers for the O_3

15 measurements would be relatively straightforward using 12 receivers each measuring both polarisations, mounted singly or with two or three LNBs bundled together at each antenna focus point. Atmospheric observations recorded by each spectrometer could then be combined to achieve a $\sqrt{24}$ = ~4.9 improvement in signal-to-noise. For OH, the greater complexity and cost of constructing 80 independent spectrometers operating at 13.441 GHz could make use of a single low temperature receiver the better option. For low temperature operation, liquid nitrogen-cooled cryostats readily attain ~90 K, meeting the

20 requirements for O_3 observations. However, this LNB cooling method requires a supply of liquid nitrogen into either a continuous flow or reservoir design of cryostat. Alternatively, LNBs mounted on the cold-head of a Gifford-McMahon closed-cycle helium cryocooler (e.g., Radebaugh, 2009; de Waele, 2011) could be operated continuously at temperatures in the range 4–77 K.

The next steps towards developing suitable instruments include selecting and testing commercially-available LNB receiver

25 and antenna components and verifying the optimum configuration for making the proposed mesospheric O_3 and OH observations. If tests confirm that low-cost satellite TV components can reliably achieve the desired performance (e.g., LNBs with noise factor of 0.23 dB or lower) then the increased complexity and cost of designing and building a system using numerous multiple receivers or cryogenic cooling would be avoided, for O_3 11.072 GHz measurements at least. It should be noted that other natural, and man-made, sources may produce signals at frequencies that overlap the O_3 and OH microwave

30 emissions. Potential interference from such sources may be mitigated by making observationsobserving at remote, high-latitude or polar locations and by carefully pointing of the receiver antenna to minimise directional pick-up of spurious signals in the instrument field-of-view. OH microwave line Zeeman splitting needs to be adequately modelled in the retrieval algorithm, and the values of the spectroscopic parameters need to be verified and optimised. Furthermore, during geomagnetic storms rapid, localised fluctuations in the Earth's magnetic field can occur that which may not be well reproduced by a priori

data from models such as IGRF. Instead, ~~it may be appropriate~~a better approach may be to use magnetometer data provided by nearby geophysical observatories to constrain the OH retrievals, or to allow the retrieval to adjust the magnetic field from a priori values to give an optimum fit to the observed line-shape.

5 Data availability

5   ~~Model~~ The model and simulation datasets (Newnham et al., 2018) used in this study are available from the UK Polar Data Centre (https://www.bas.ac.uk/data/uk-pdc/).

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Table 1. Calculated performance for ozone (O₃) 11.072 GHz and hydroxyl (OH) 13.441 GHz radiometers with 10 kHz frequency resolution.

	O₃ 11.072 GHz				OH 13.441 GHz		
Noise factor (dB)	1.3	0.7	0.7	0.7	1.2	1.2	1.2
Reference temperature (K)	290	290	290	168	290	290	46
System noise temperature (K)	101	51	51	15	92	92	15
Number of measurement channels	2	2	24	2	2	80	2
Measurement time (h)	24	24	6	6	24	6	6
1σ rms noise (K)	3.4	1.7	1.0	1.0	3.1	1.0	1.0
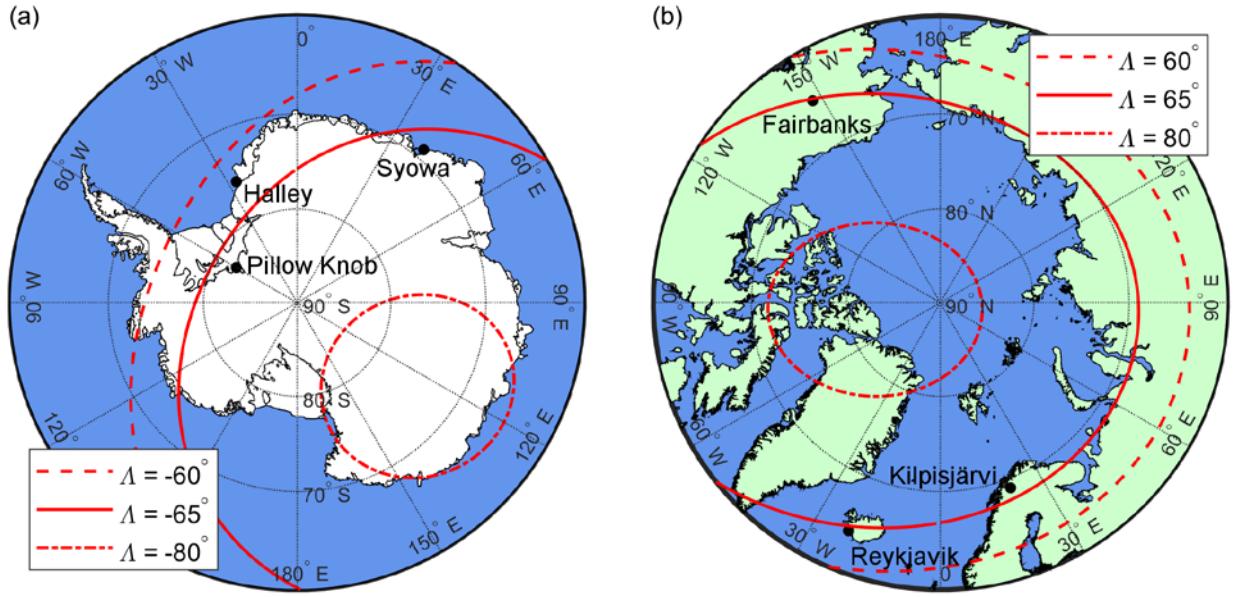
Figure 1: Maps of a) the southern hemisphere and Antarctica poleward of geographic latitude 60°S and b) the northern hemisphere and Arctic region poleward of geographic latitude 60°N. The red dashed, solid and dot-dashed lines on each map show the geomagnetic latitudes $\Lambda = \pm 60°$, $\Lambda = \pm 65°$, and $\Lambda = \pm 80°$ respectively, calculated for 1 January 2015 and an altitude of 80 km using the IGRF-12 internal field model (Thébault et al., 2015). Filled black circles indicate the locations of Halley station (75°37'S, 26°15'W, $\Lambda = -62.3°$), Syowa station (69°00'S, 39°35'E, $\Lambda = -66.6°$), and Pillow Knob refuelling station (82°30'S, 60°00'W, $\Lambda = -67.3°$) in a) and Kilpisjärvi (69°03'N, 20°49'E, $\Lambda = 66.2°$), Finland, Reykjavik (64°08'N, 21°56'W, $\Lambda = 64.4°$), Iceland, and Fairbanks (64°51'N, 147°43'W, $\Lambda = 65.2°$), Alaska in b).
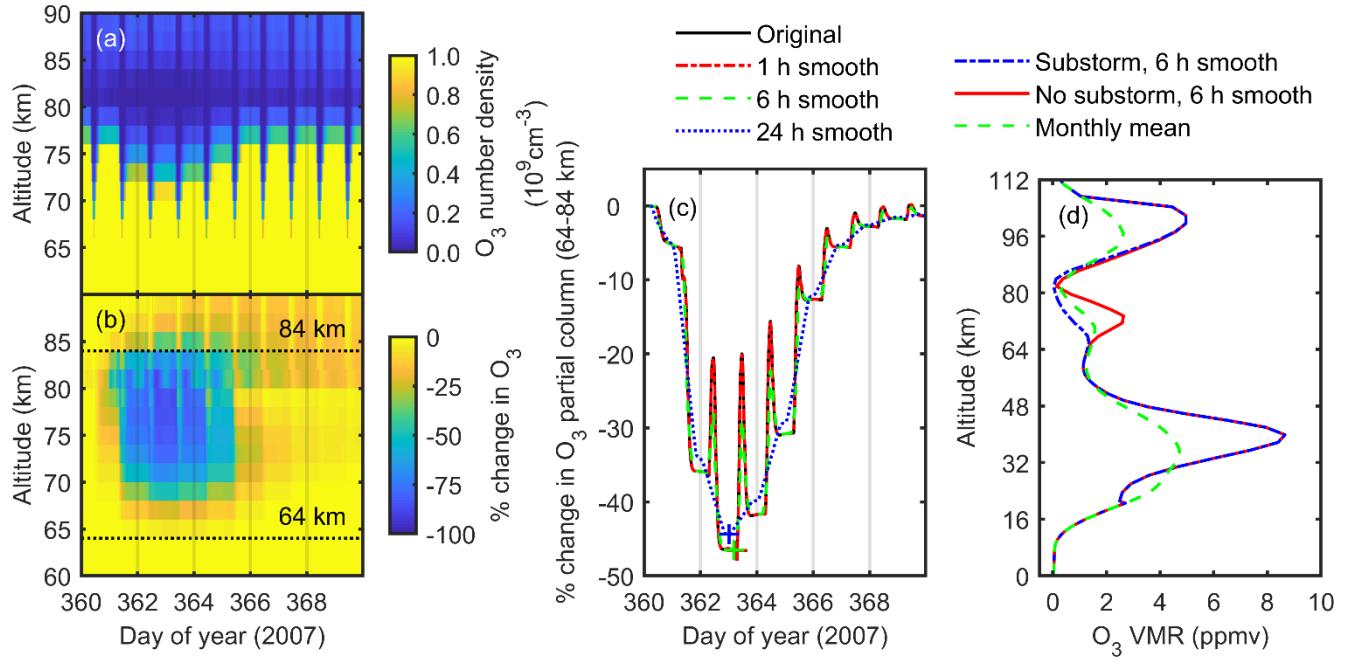
Figure 2: Derivation of ozone (O_3) volume mixing ratio profiles from atmospheric model data-sets. Panel a) shows O_3 number densities calculated by the SIC model for December 2007 sub-storm conditions at Kilpisjärvi (69°03' N, 20°48' E), Finland. Panel b) shows the percentage change in modelled O_3 abundance from background (no sub-storm) conditions. Panel c) shows the percentage change in O_3 partial column over altitudes 64–84 km during sub-storm conditions for the original (15 min) model resolution and 1 h, 6 h, and 24 h smoothed data. '+' symbols indicate the largest decreases in O_3 partial column at each nominal time resolution. Plot d) shows the 6 h-smoothed O_3 VMR profiles for December sub-storm, background (no sub-storm), and monthly mean conditions.
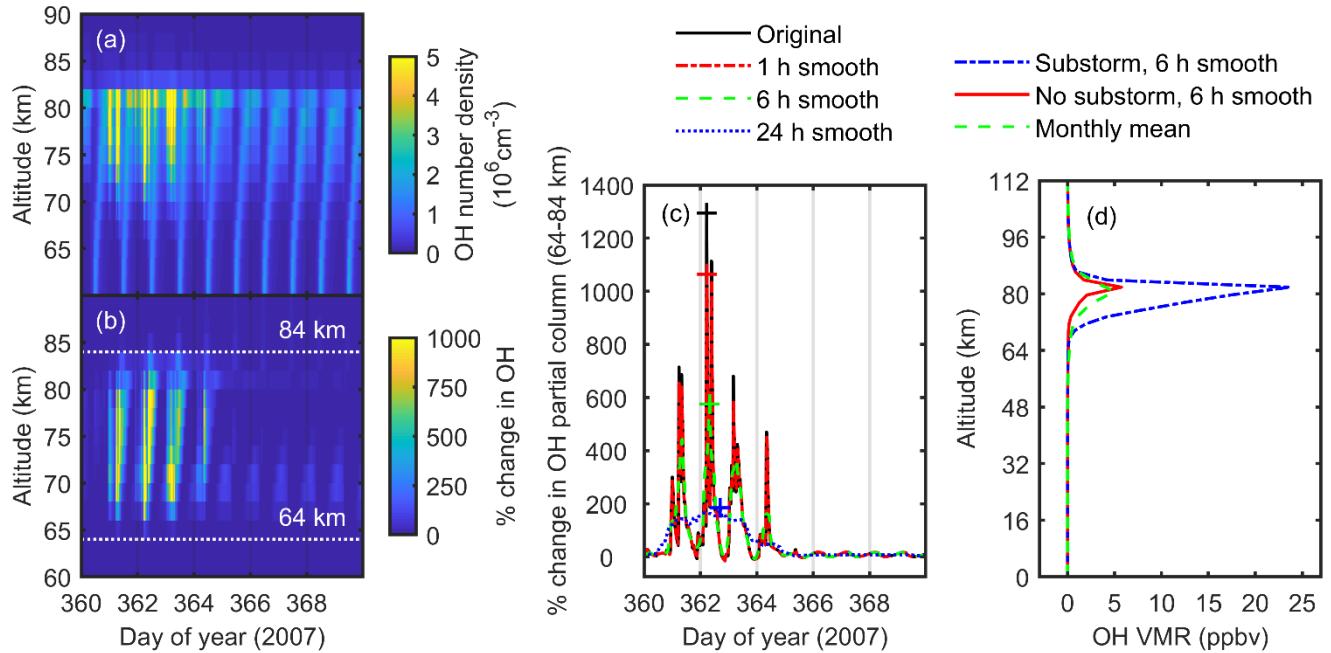
Figure 3: Derivation of hydroxyl radical (OH) volume mixing ratio profiles from atmospheric model data-sets. Panel a) shows OH number densities calculated by the SIC model for December 2007 sub-storm conditions at Kilpisjärvi (69°03' N, 20°48' E), Finland. Panel b) shows the percentage change in modelled O_3 abundance from background (no sub-storm) conditions. Panel c) shows the percentage change in OH partial column over altitudes 64–84 km during sub-storm conditions for the original (15 min) model resolution and 1 h, 6 h, and 24 h smoothed data. '+' symbols indicate the largest increases in OH partial column at each nominal time resolution. Plot d) shows the 6 h-smoothed OH VMR profiles for December sub-storm, background (no sub-storm), and monthly mean conditions.
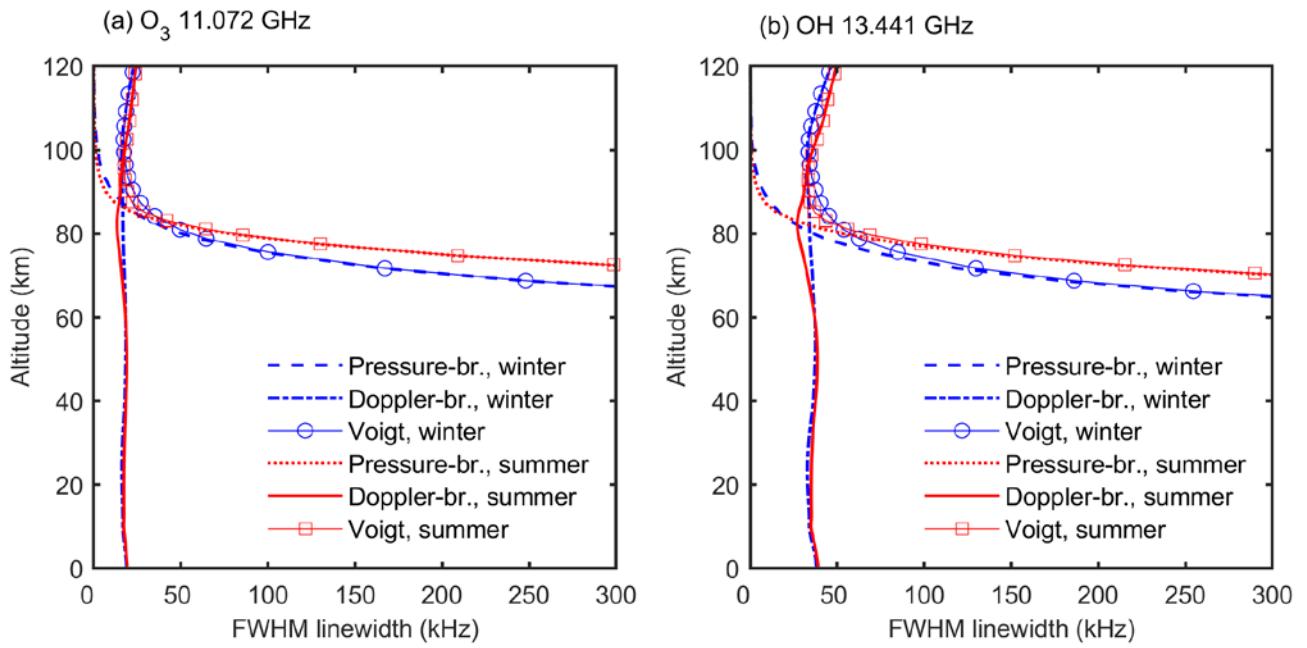
Figure 4: Pressure-broadened, Doppler-broadened, and Voigt full-width half-maxima (FWHM) linewidths for a) ozone (O₃) 11.072 GHz and b) OH 13.441 GHz emission lines, calculated for mean winter (DJF) and mean summer (JJA) conditions at Kilpisjärvi (69°03' N, 20°48' E), Finland.
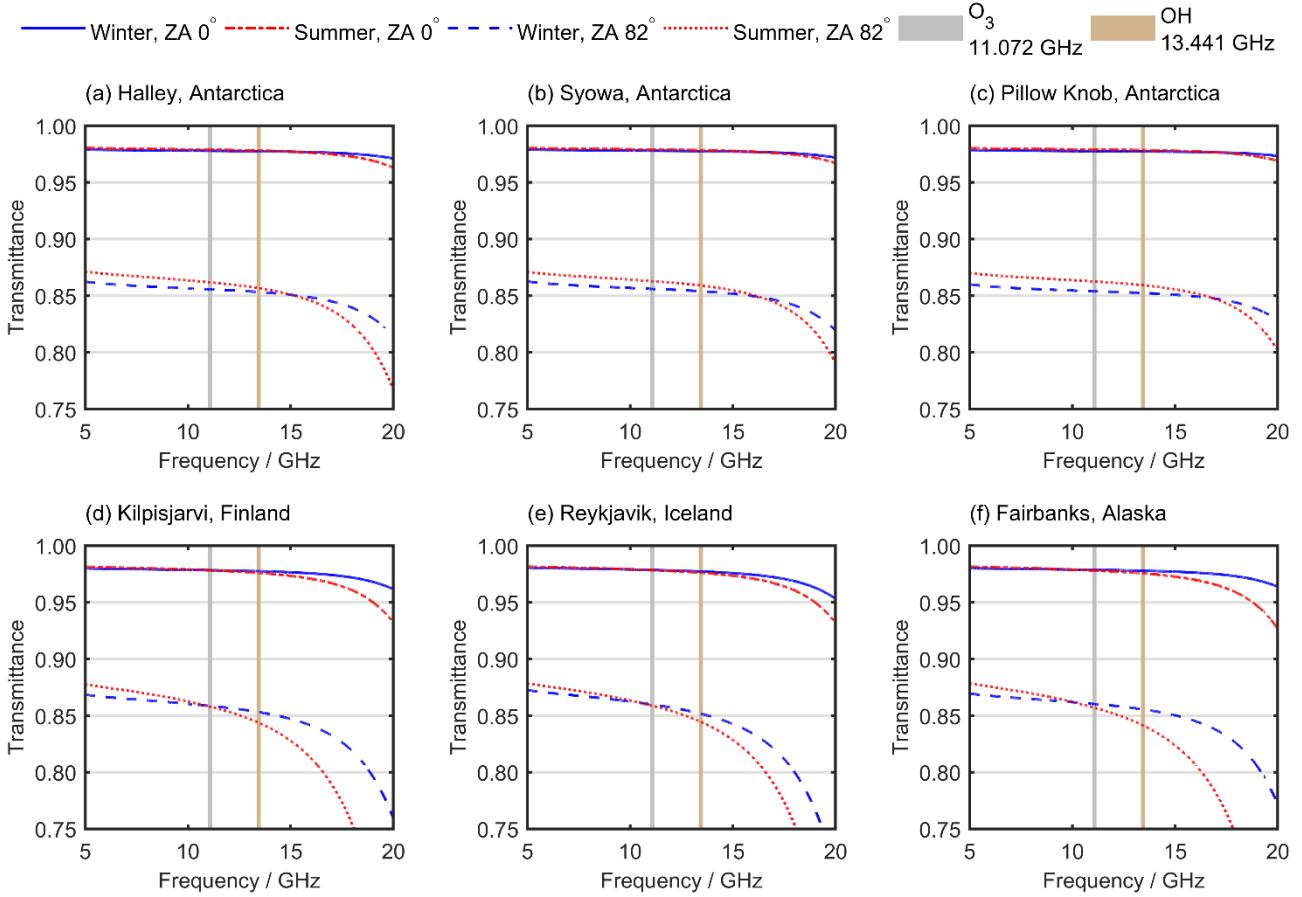
Figure 5: Survey atmospheric transmittance spectra at six polar locations for the frequency range 5–20 GHz, calculated on a 1 MHz frequency grid. In plots a)–c) transmittances calculated at zenith angle (ZA) 0° (i.e. viewing vertically upwards) and 82° are shown for mean summer (DJF) and mean winter (JJA) conditions at three Antarctic locations. In plots d)–f) transmittances calculated at ZA 0° and 82° are shown for mean summer (JJA) and mean winter (DJF) conditions at three Arctic locations. The light grey and light brown vertical lines indicate the frequencies of the ozone (O_3, 11.072 GHz) and hydroxyl (OH, 13.441 GHz) emission lines.
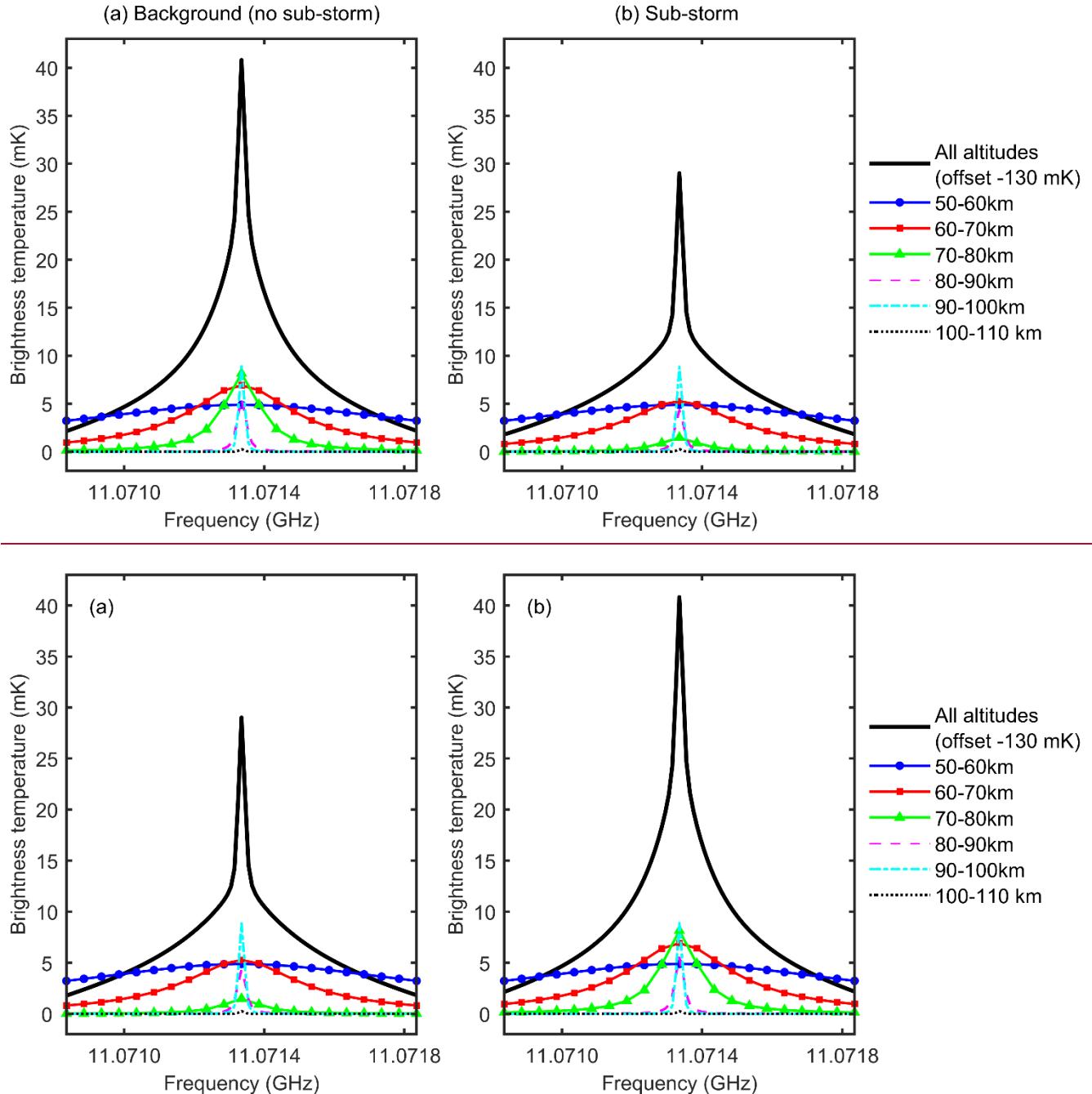
Figure 6: Simulated atmospheric spectra for the ozone (O₃) 11.072 GHz emission line in clear-sky, December conditions using a ground-based radiometer with 10 kHz frequency resolution, 82° zenith angle, and 0° azimuthal angle located at Kilpisjärvi (69°03' N, 20°48' E), Finland. Solid black curves show the overall OH emission from all altitudes. Coloured lines show the

contributions from 10 km altitude intervals in the range 50–110 km. Plots a) and b) show the spectra for background (no sub-storm) ~~sub-storm~~ and sub-storm ~~background (no sub-storm)~~ conditions respectively.
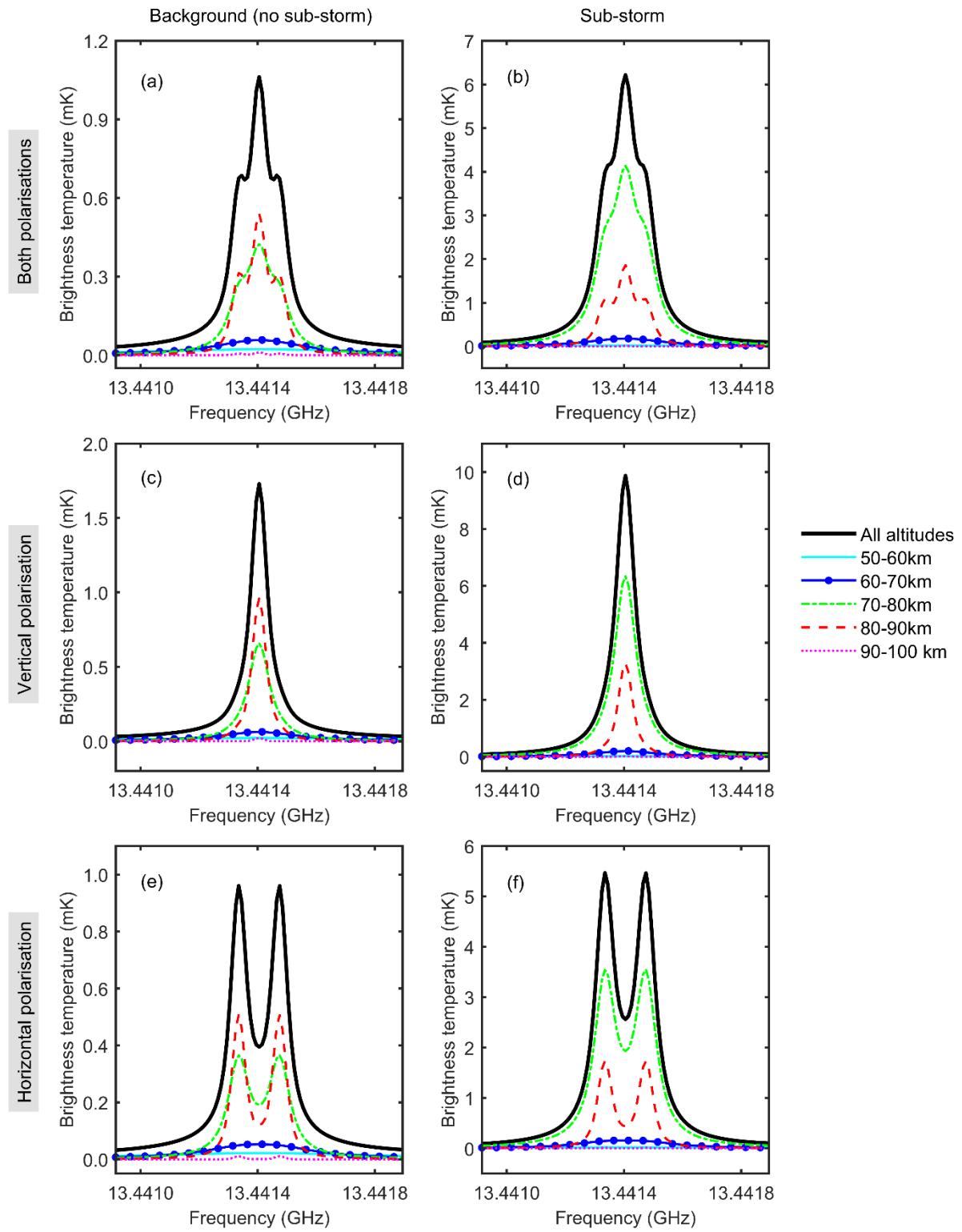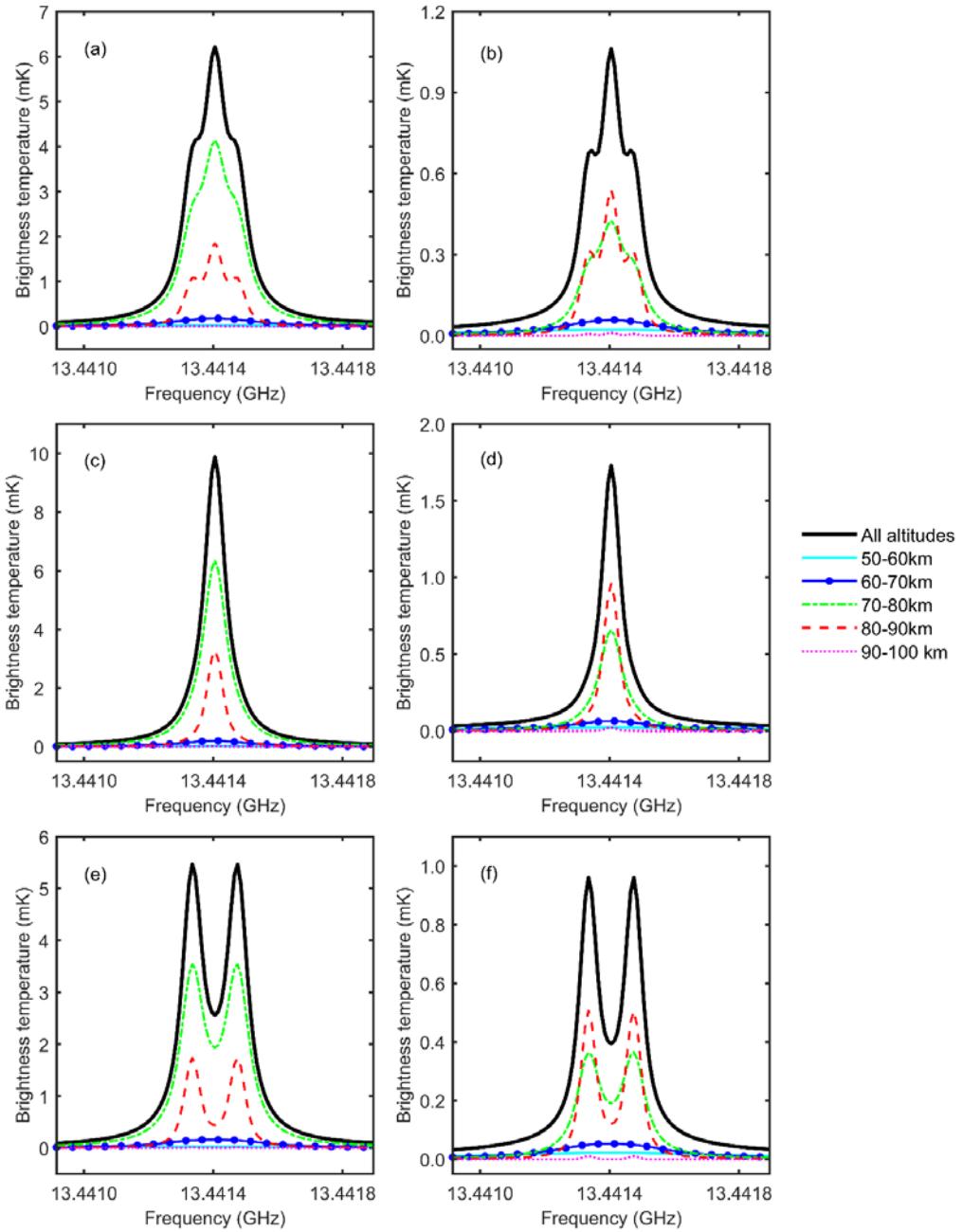
Figure 7: Simulated atmospheric spectra for the hydroxyl (OH) 13.441 GHz emission line in clear-sky, December conditions using a ground-based radiometer with 10 kHz frequency resolution, 82° zenith angle, and 0° azimuthal angle located at Kilpisjärvi (69°03' N, 20°48' E), Finland. Contributions from other atmospheric species have been removed to show changes in the brightness temperature spectra due to OH. Solid black curves show the overall OH emission from all altitudes. Coloured lines show the contributions from 10 km altitude intervals in the range 50–100 km. Plots a) and b) are for all sensor polarisations in ~~background (no sub-storm) and~~ sub-storm ~~and background (no sub-storm)~~ conditions respectively. Similarly, c) and d) are for vertical sensor polarisation and e) and f) are for horizontal sensor polarisation.
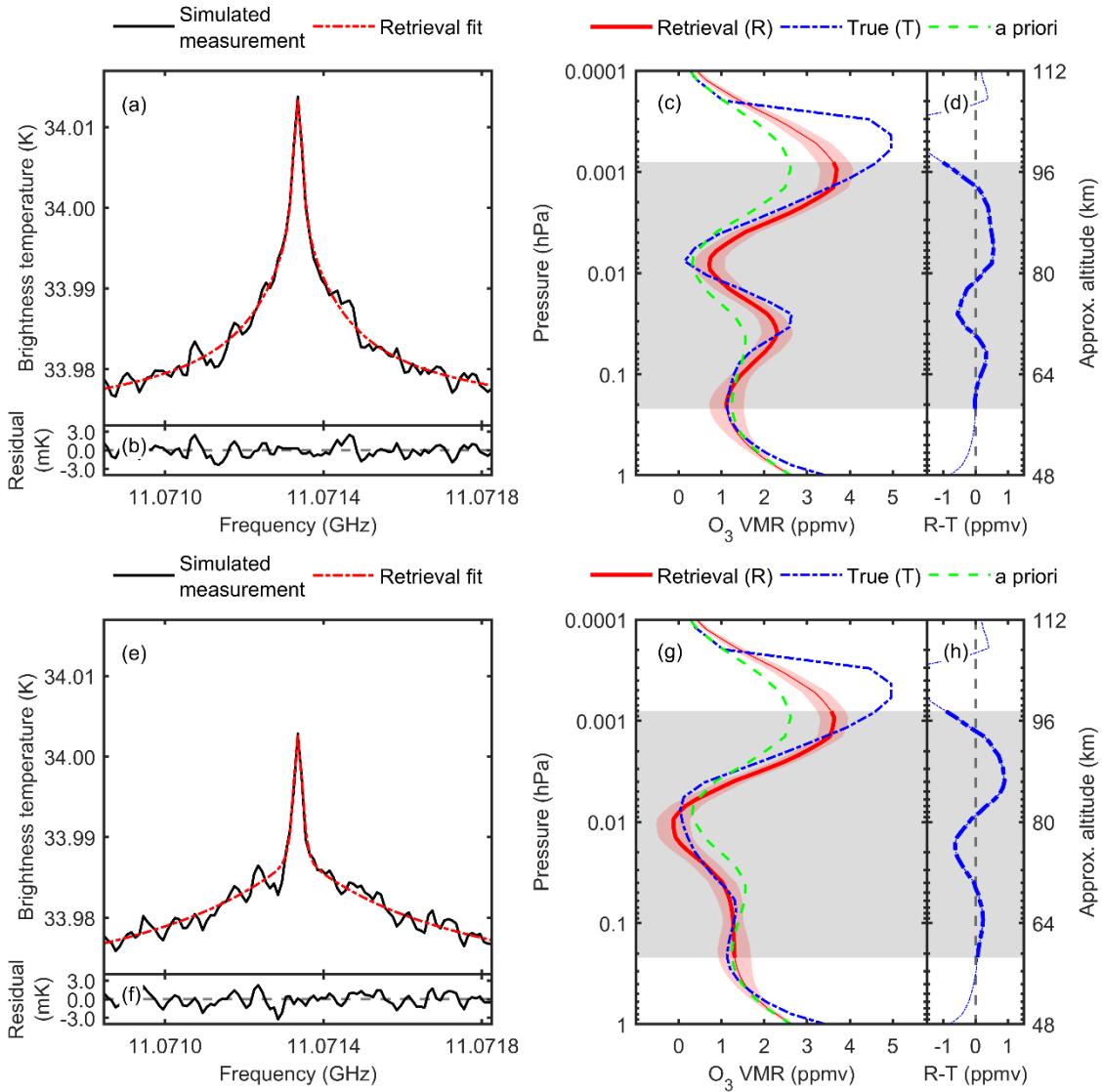
Figure 8: Ozone (O₃) retrievals for simulated 6 h observations of the O₃ 11.072 GHz emission line in clear-sky December
conditions using a ground-based radiometer at Kilpisjärvi (69°03' N, 20°48' E), Finland. The forward model clear-sky spectra
are calculated using the model O₃ profiles for December 2007 with 1 mK baseline noise, 10 kHz frequency resolution, and 82°
zenith angle. a)–d) show the results using background (no sub-storm) O₃ in the simulations, and e)–g) show the results for
sub-storm O₃. The simulated O₃ spectra and retrieval fits are shown in a) and e), and the residual differences in b) and f). The
a priori, true, and retrieved O₃ volume mixing ratio profiles, and the differences between the retrieved and true profiles, are
shown in c)–d) and g)–h) for the background and sub-storm cases respectively.
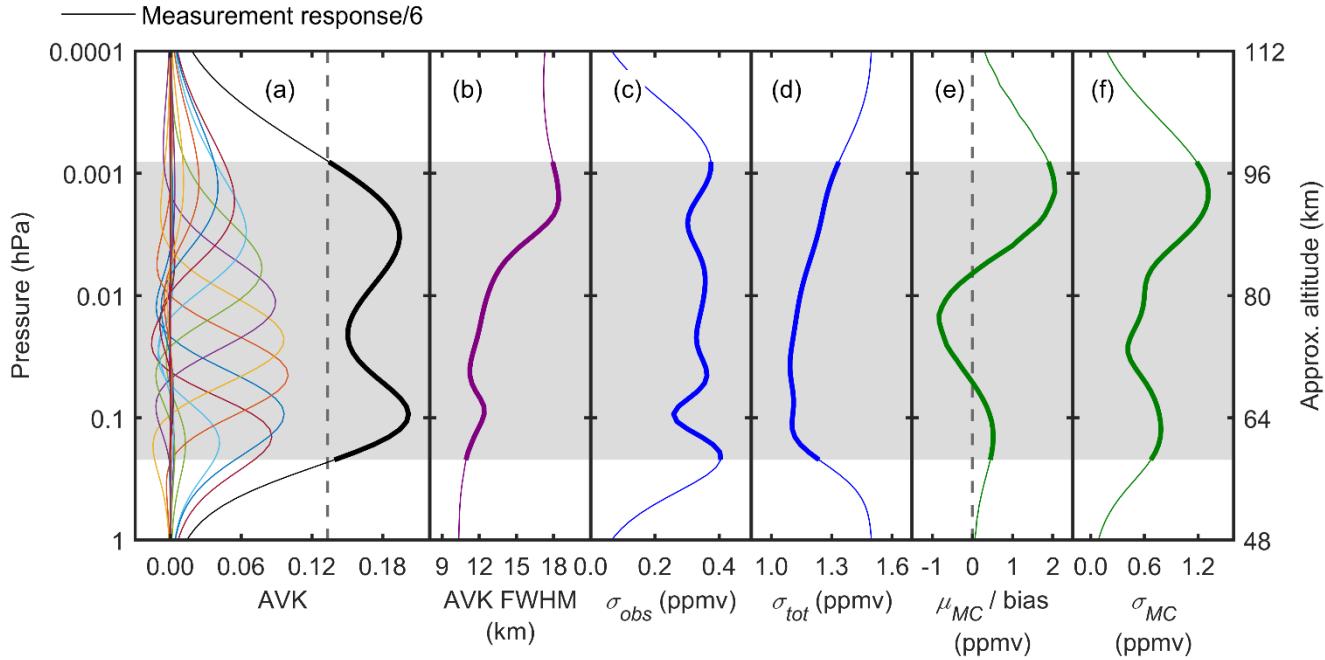
Figure 9: Ozone (O_3) retrieval diagnostics for simulated 6 h observations of the O_3 11.072 GHz emission line using a ground-based radiometer with 1 mK baseline noise level, 10 kHz frequency resolution, and 82° zenith angle located at Kilpisjärvi (69°03' N, 20°48' E), Finland in clear-sky December conditions. In a) every sixth averaging kernel, and the scaled measurement response (MR), are shown. Panel b) shows the full-width half maxima of each averaging kernel (AVK FWHM). The measurement uncertainty (σ_{obs}) and total uncertainty (σ_{tot}) are shown in c) and d) respectively. The mean (μ_{MC}) and standard deviation (σ_{MC}) of the differences between the retrieved and "true" profiles from Monte Carlo (MC) error analysis using 500 repeat inversions are shown in e) and f) respectively. The vertical grey dashed line in a) shows the cut-off for MR ≥ 0.8. The grey shaded regions and the thicker sections of the plotted curves indicate the pressure / altitude ranges where MR ≥ 0.8.

Figure 10: Rows of the Jacobian describing the ozone (O_3) retrieval, normalised by the layer thickness of the retrieval grid. The data are for retrievals from simulated 6 h observations of the O_3 11.072 GHz emission line using a ground-based radiometer with 1 mK baseline noise level, 10 kHz frequency resolution, and 82° zenith angle located at Kilpisjärvi (69°03' N,

20°48' E), Finland in clear-sky December conditions. Rows of the Jacobian matrix for selected altitude levels are plotted in a). The grey scale in b) indicates the values of the Jacobian matrix.
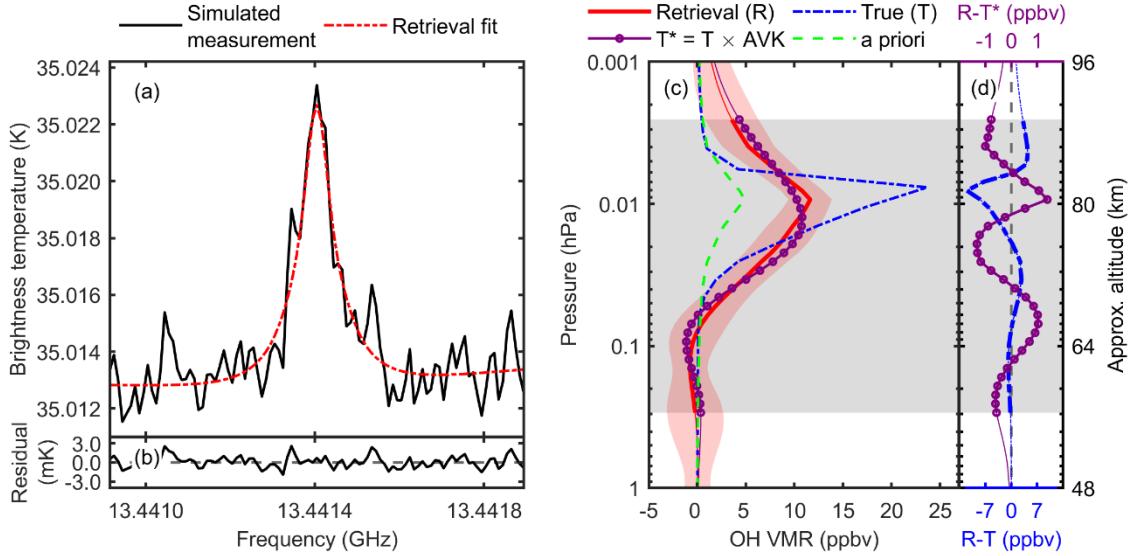
Figure 11: Hydroxyl (OH) retrievals for simulated 6 h observations of the OH 13.441 GHz emission line using a ground-based radiometer with vertical sensor polarisation at Kilpisjärvi (69°03' N, 20°48' E), Finland. The forward model clear-sky spectrum is calculated using the model OH profile for substorm conditions in December 2007 with 1 mK baseline noise, 10 kHz frequency resolution, 82° zenith angle, and 0° azimuthal angle (i.e. north-pointing). The simulated OH spectra and retrieval fit are shown in a), and the residual differences in b). The a priori, true, and retrieved OH volume mixing ratio profiles, and the differences between the retrieved and true profiles, are shown in c) and d). Note that the scales differ for the upper and lower axes of d).
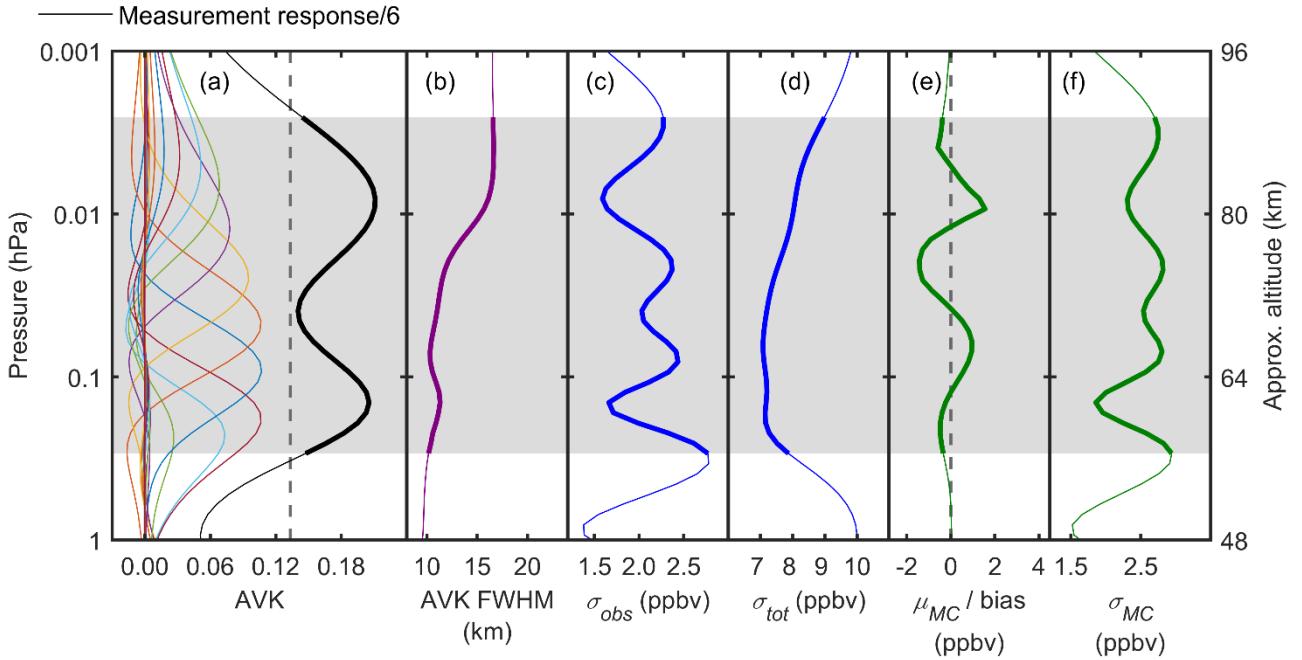
Figure 12: Hydroxyl (OH) retrieval diagnostics for simulated 6 h observations of the OH 13.441 GHz emission line using a ground-based radiometer with vertical polarisation, 1 mK baseline noise level, 10 kHz frequency resolution, 82° zenith angle, and 0° azimuthal angle (i.e. north-pointing) located at Kilpisjärvi (69°03' N, 20°48' E), Finland in clear-sky December conditions. In a) every sixth averaging kernel, and the scaled measurement response (MR), are shown. Panel b) shows the full-width half maxima of each averaging kernel (AVK FWHM). The measurement uncertainty (σ_{obs}) and total uncertainty (σ_{tot}) are shown in c) and d) respectively. The mean (μ_{MC}) and standard deviation (σ_{MC}) of the differences between the retrieved and "true" profiles from Monte Carlo (MC) error analysis using 500 repeat inversions are shown in e) and f) respectively. The vertical grey dashed line in a) shows the cut-off for MR ≥ 0.8. The grey shaded regions and the thicker sections of the plotted curves indicate the pressure / altitude ranges where MR ≥ 0.8.

Figure 13: Rows of the Jacobian describing the hydroxyl (OH) retrieval, normalised by the layer thickness of the retrieval grid. The data are for retrievals from simulated 6 h observations of the OH 13.441 GHz emission line using a ground-based radiometer with vertical polarisation, 1 mK baseline noise level, 10 kHz frequency resolution, 82° zenith angle, and 0° azimuthal angle (i.e. north-pointing) located at Kilpisjärvi (69°03' N, 20°48' E), Finland in clear-sky December conditions.

Rows of the Jacobian matrix for selected altitude levels are plotted in a). The grey scale in b) indicates the values of the Jacobian matrix.