Answer to Anonymous Referee #1 Received and published: 18 April 2019

General comments:

This manuscript deals with an interesting effect not properly addressed by previous studies on the population of rotational levels of hydroxyl radicals in the mesosphere / lower thermosphere region of the Earth's atmosphere. The simple fact that kinetic temperature varies within the altitude range of the OH emission layer (for a wave or tidal perturbation possibly dramatically) will lead to an apparent deviation of the population of higher rotational states from LTE in ground-based observations. While this effect may not explain the majority of observed non-thermal populations of higher rotational levels, it certainly is an interesting effect that should be reported and considered in future studies. For this reason this manuscript is of interest to the aeronomy community and should eventually be published in my opinion. I ask the authors to consider the (mainly minor) comments below.

We thank the referee for all comments and helpful feedback. We address each comment in turn below.

Specific comments:

Page 3, line 15: "from the empirical model of the US Naval Research Laboratory Mass Spectrometer and Incoherent Scatter radar (NRLMSISE-00)" sounds a little odd and is misleading. I suggest something like: "from the US Naval Research Laboratory's Mass Spectrometer and Incoherent Scatter radar model (NRLMSISE-00)"

The text has been changed as suggested.

Page 4, line 1: "The temperature dependent rate coefficients for THESE reactions" Should this perhaps be ".. for THIS reaction"? The statement seems to refer to reaction (2) only.

The text has been changed as suggested.

Page 5, line 16: "Where A_w(z) is a function of altitude" Please provide more information on the altitude dependence of the wave amplitude, perhaps show a plot.

We follow here the wave breaking mechanism of (Holton, 1982). To clarify the amplitude behaviour with altitude, we have added the following passage and figure to the text:

"Where φ_w is the wave's phase, and $A_w(z)$ is a function of altitude so as not to exceed the dry adiabatic lapse rate. The lower edge of the model is 74 km altitude. The wave amplitude as a function of altitude is shown in Figure 2 for the case of a wave with an amplitude of 10 K at 74 km altitude and a vertical wavelength of 15 km. This example is given for an isothermal atmosphere of 200 K (Figure 2 (a), dashed-black). The wave grows in amplitude with altitude to conserve energy (dashed-blue), but at regions where the lapse rate exceeds the dry adiabatic lapse rate (here between 90 and 95 km, and between 110 and 115 km), the wave loses energy and the amplitude decreases (Holton, 1982). The breaking wave is shown in red in Figure 2 (a). Figure 2 (b) shows the instantaneous lapse rate (change in temperature with altitude) of the non-breaking wave (dashedblue) and the breaking wave (red), which never crosses the dry adiabatic lapse rate of -10 K/km (dashed-black). Figure 2 (c) shows the amplitude of the non-breaking wave (dashedblue) which increases exponentially and for the breaking wave (red), which decreases at the altitudes where the wave dissipates energy.



Figure 2: (a) Example of a wave of amplitude 10 K at 74 km altitude perturbing an isothermal atmosphere at T = 200 K (dotted-black). The dashed-blue line represents the wave without any breaking, and the red line is the breaking wave as used in the model. (b) The rate of change of temperature with altitude for the wave shown in (a). When the temperature changes faster than -10 K/km (dashed-black) the wave breaks. (c) At these altitudes, the amplitude of the breaking wave (red) decreases, while the non-breaking wave continues to grow exponentially (dashed-blue)."

Page 5, same line: "and \$nphi_w\$ for this example is defined as zero at an altitude of 74 km" This statement suggests that the phase is altitude dependent. Is this really the case? I assume it is constant – then the reference to a specific altitude (74 km) can be removed.

The phase shift \$nphi_w\$ is not altitude dependent, as the referee said. The text has been changed to be less confusing. It now reads:

"Where φ_w is the wave's phase (constant with altitude), and $A_w(z)$ is a function of altitude so as not to exceed the dry adiabatic lapse rate. The lower edge of the model is 74 km altitude."

Page 5, same paragraph: It would be good to state here already, whether the assumed wave perturbation may really occur in the atmosphere, or whether this is an extreme case that essentially never occurs.

We agree that is it good to state this already here. We have added the following sentence to the text.

"Waves with similar amplitudes have been observed at these altitudes (Picard et al., 2004)".

Page 6, line 12: "Where N_v ' is the integral of the vibrational band VER" Is this really the case? I think N_v ' is not an integrated VER, but rather the total (integrated) population of the v' level. Otherwise the units don't fit.

 N_v' is indeed the integral over the population, or in terms of VER, you can also put the integral as $N_v(z) \cdot \omega_{v'v'} / \tau_{v'}$. We clarified this in the text. It now reads:

"Where N_{v} is the integral of the vibrational population $N_{v'}(z)$ over altitude, and T is the effective rotational temperature of the altitude-integrated spectrum."

Page 7, Figure 3, upper abscissa legend: Please add a space between "1.5" and "2.5"

We have rotated the labels by 90 degrees to increase readability. The new figure now looks like this:



Page 8, line 7: "the original background profile is retrieved" I suggest replacing "retrieved" by "obtained". For me as a "retrieval person" "retrieved" here means the temperature retrieval using the OH technique."

The text has been changed as suggested.

Page 8, line 8: only a minor comment, but "higher and lower temperatures" is perhaps more accurate than "warmer and colder temperatures", because temperature cannot really be warm/cold. I leave it up to the authors to decide, whether they want to change this.

The text has been changed as suggested.

Page 9, Figure 4: It would be good to separate the two panels a little, add some space between panel a and b.

We have added some space between these two panels. The figure now looks like this:



Figure 5: The calculated apparent excess population of the OH (7,4) P(J') lines relative to the Boltzmann population of the fitted temperature to the lowest three lines. Red on the left is calculated with the climatological temperature gradient shown from NRLMSISE-00 above Boulder, Colorado (40.0° N; 105.6° W) in mid-July. Blue on the right is calculated with a wave of $A_w = 30$ K at 90 km, $\lambda_w = 30$ km and a phase that yields the maximum effect, $\varphi_w = 4.9$ rad. This is the same wave as presented in Figure 4. The distribution has a $\beta = 1.9$ %. The grey bars represent the measurements ascribed to NLTE effects from Pendleton et al. (1993)."

Page 9, line 18: "The first three rotational levels reproduce the same population as an atmosphere in LTE characterised by a single temperature" It's not entirely clear, whether this statement refers to the results of Pendleton et al. (1993). I think this is the case and suggest stating this explicitly.

This statement refers to both this paper and the one of Pendleton et al. (1993). We have changed this sentence to clarify this. It now reads:

"The first three rotational levels in this study and in Pendleton et al. (1993) have populations that follow a Boltzmann distribution characterized by a single temperature, and above that, there is excess population."

Page 10, line 7: "In Fig. 5(a) the non-linearity of the temperature fit increases with both wave strength and vertical wavelength" Looking at the Figure, this statement is not generally correct, is it? There are different regimes showing different behavior, e.g., for a vertical wavelength of 10 km, beta decreases with increasing amplitude. Please rephrase this sentence.

The referee is correct. We have clarified this in the text. It now reads:

"Figure 6 (a) shows that for vertical wavelengths above about 20 km the non-linearity of the temperature fit increases with both wave strength and vertical wavelength approaching 2% of the linear temperature variation (see Equation 10). For shorter vertical wavelengths, the non-linearity is smaller (at about 0.5 %) and decreases with increasing wave amplitude."

Page 10, line 9: "Figure 5 (b) shows the phase that yields the highest : : :" The Figure does not show the phase, but the apparent excess population for the phase with : : : Please rephrase.

The figure shows the apparent excess population for a wave with a given wavelength and amplitude using the phase that yields the greatest effect. We have now clarified this in the text, which reads:

"Figure 6 (b) shows the apparent excess populations for a wave with a phase that yields the highest apparent excess population for a given wave amplitude and wavelength."

Page 11, line 10: "Instead it is due to the" It's not clear, what "it" refers to in this sentence. I assume it is "the difference in the apparent excess population" mentioned a few sentences above. Please state it explicitly.

The "it" refers to the difference in the apparent excess population between the (3,1) and (7,4) bands. This has been clarified and the sentence now reads:

"Instead, the difference in the apparent excess population between the (3,1) and (7,4) bands is likely to be due to the compressed rotational energy structure of the higher vibrational levels that lie closer to the dissociation limit."

Page 12, line 9: "The simulations executed here show that these temperature profiles can change the populations of the different rotational lines" I have two comments on this sentence: 1) it is not surprising that temperature affects the rotational population of a vibrational state and I think this is not what you actually intend to say. The important point is that the vertical temperature variation leads to an apparent non-thermal population for an observer on the ground, right? 2) can "rotational lines" be populated? The rotational "states" or "levels" are populated and they give rise to the emission.

We agree with the referee on both points. We have changed the sentence to clarify the apparent non-thermal population and exchanged "line" with "level". The whole sentence now reads:

"The simulations executed here show that these temperature profiles can create an apparent non-thermal population of the rotational levels in a given Meinel (v', v'') band."

Typos etc.:

General: The section titles are all upper cases, which – I believe – is not the ACP standard.

The section titles have been changed as suggested.

Page 1, reaction (1): The "-" sign inside the parentheses may be interpreted as a minus sign and I think it is not really necessary.

The minus sign was supposed to indicate that this energy is the excitation of the OH^{*}. We agree however that it might be confusing and have deleted this sign.

Page 3, caption of Figure 1, line 1: "red- dashed" -> "red-dashed" ?

The text has been changed as suggested

Page 4, line 3: "Loss processes include losses DUE to O" ?

The text has been changed as suggested

References:

Holton, J. R.: The Role of Gravity Wave Induced Drag and Diffusion in the Momentum Budget of the Mesosphere, J Atmos Sci, 39, 791-799, 1982. Pendleton, W. R., Espy, P. J., and Hammond, M. R.: Evidence for non-local-thermodynamic-equilibrium rotation in the OH nightglow, J Geophys Res-Space, 98, 11567-11579, 1993. Picard, R. H., Wintersteiner, P. P., Winick, J. R., Mertens, C. J., Mlynczak, M. G., III, J. M. R., Gordley, L. L., Ward, W. E., She, C. Y., and O'Neil, R. R.: Tidal and layer structure in the mesosphere and lower thermosphere from TIMED/SABER CO2 15- μ m emission, SPIE, 2004.