

Answer to Anonymous Referee #2

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General comments:

This study focusses on the impact of kinetic temperature gradients in the OH airglow emission layer on column-integrated rotational level population distributions. As the populations of high rotational levels tend to originate from altitudes with higher kinetic temperatures than those related to low rotational levels, the rotational level population distribution of a fixed vibrational level cannot be described by a single temperature. The temperature tends to increase with rotational level, which looks like a non-LTE effect. The authors simulated different wave-perturbed temperature profiles combined with a chemical model for the OH emission layer (depending on the vibrational level) to estimate the magnitude of the effect. A comparison with measured OH populations from Pendleton et al. (1993) then showed that this pseudo-non-LTE effect can significantly contribute to the apparent non-LTE deviations in the rotational level population distribution if the wave amplitudes are large.

There has not been a detailed study of the investigated effect in the literature, so far. Hence, it is justified to publish a paper on this topic, although I guess that the effect appears to be negligible in most cases. As the study does not discuss the distribution of true temperature gradients (either from satellite or lidar data), I would appreciate an extension of the analysis in order to better understand the relevance for observed rotational level population distributions like those from Pendleton et al. (1993) or more recent studies. For OH-based estimates of the kinetic temperature, rotational temperatures are usually derived from the first three or four rotational lines. Hence, it would also be interesting to know whether a noteworthy contribution of the discussed effect is possible. OH modelling results in the literature should not be affected as such models are calculated altitude-dependent. Only the interpretation of the column-integrated populations may change slightly.

Apart from the desired clarifications concerning the impact of the effect, the quality of the discussion needs to be significantly improved. In particular, due to the lack of precision in the use of scientific terms, the paper is often confusing and misleading (see detailed comments). Thus, this paper needs a major revision to be acceptable for publication in ACP.

We thank the referee for their extensive and helpful comments.

This study uses a temperature profile from the reanalysis NRLMSIS-00, which is based on “realistic temperature gradients”. We have also been careful to ensure that the waves we present have been reported in the literature using a variety of observational techniques and are therefore physical (Hagan et al., 1999; Oberheide et al., 2011; Picard et al., 2004; She et al., 2002; Shepherd and Fricke-Begemann, 2004; Zhang et al., 2006). The results presented here describe the temperature gradients that arise over the range of these observed waves. We believe that a systematic scan of different waves spanning the range of temperature gradients arising from observed waves presents a stronger argument than a calculation of a specific observation from satellite or lidar, each of which has specific observational bias.

The referee is right that OH temperatures are typically derived from lines with $N < 5$, which are the first three (for the P-branch) or four (for the Q- and R-branches) lines. These lines are not affected, or are only marginally affected, by the findings presented in this paper as is clearly stated in the manuscript. The focus of this paper is not to argue an impact on the majority of OH temperature

calculations, but rather to highlight the impact on attempts to quantify other contributions to the excess populations observed in the higher lying rotational levels.

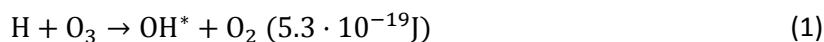
We have responded to all comments in order below and hope that the scientific precision is now acceptable to the referee and that we have cleared up any confusing or misleading text. Again, we thank the referee for their comments and for their help in improving the quality of the paper.

Specific comments:

P.1, L.23: "kcal" is an old-fashioned physical unit. The SI equivalent would be "KJ".

We deleted the kcal unit and use kJ now instead. The whole passage now reads:

"The 8-km thick airglow layer is created at about 90 km altitude (Baker and Stair, 1988; Xu et al., 2012) by the highly exothermic reduction of ozone:



The excess heat of reaction, $\sim 5.3 \cdot 10^{-19}\text{J}$, produces the OH^ in excited vibrational quantum levels of $v'=6-9$ (e.g. Mlynczak and Solomon (1993))."*

P.1, L.29-30: The rotational temperature fits for the OH nascent populations used by Dodd et al. (1994) originate from Llewellyn & Long (1978).

The $v' = 9$ temperature is indeed from Llewellyn & Long (1978), while the $v' = 6$ temperature is from Dodd et al. (1994). This has been corrected. The text now reads:

"Llewellyn & Long (1978) reported a nascent temperature of 760 K for $v'=9$, whereas others report temperatures as high as 9000-10000 K (Dodd et al, 1993; Oliva et al., 2015; Kalogerakis et al., 2018)."

P.1, L.30-31: Khomich et al. (2008) do not report rotational temperatures of about 10,000 K. They refer to such values only in the context of vibrational temperatures. Nevertheless, rotational temperatures of this magnitude have already been measured (Oliva et al. 2015, Kalogerakis et al. 2018).

We have changed the references here as suggested.

P.1, L.32 - P.2, L1: In general, it cannot be stated that low rotational levels (the rotational quantum numbers could be mentioned) are in LTE. In the case of very low vibrational levels, this might be close to reality, but for higher vibrational levels, there appears to be a significant excess even for the lowest states (e.g. Noll et al. 2018). The problem is that the full thermalisation is not achieved due to an insufficient number of thermalizing collisions in the especially short lifetimes of the high vibrational levels. Without the vibrational level changes by collisions and photon radiation, LTE would be possible.

We use the term LTE loosely in the sense that the distribution of radiation from the levels can be describes by a single temperature Boltzmann distribution. For low rotational levels $N \leq 4$, there is evidence that this temperature is consistent with the atmospheric temperature (Oberheide et al., 2006; von Zahn et al., 1987). As such, we have taken the approach that at each altitude, the temperature characterizing the Boltzmann distribution is the local atmospheric temperature, and examine only the deviations to that temperature caused by integrating each rotational line intensity through the temperature gradients across the layer.

We have made this passage clearer, adding specifically which levels we are discussing and that the studies referenced find a single temperature describes the Boltzmann distribution. We have also added a reference to the paper of Noll et al. at the end of this paragraph. Since our study however uses strict LTE conditions in the model and has no NLTE effects programmed in the model, this paragraph remains quite short, since its intention is to give an overview of the general research done in the field rather than to discuss or make inferences on the impact of NLTE effects on the lower rotational levels.

The new paragraph now reads:

“Low rotational levels (with $N \leq 4$) with energy separations less than kT , the amount typically exchanged during collisions, have been observed to have efficient energy transfer in the thermalisation process (Maylotte et al., 1972; Polanyi and Sloan, 1975; Polanyi and Woodall, 1972). Thus, emission from these states has been observed to be characterised by a single temperature Boltzmann distribution (Harrison et al., 1971; Harrison et al., 1970; Pendleton et al., 1993; Perminov et al., 2007; Sivjee et al., 1972; Sivjee and Hamwey, 1987). However, emission observed from the higher rotational levels ($N > 4$), where the energy separation exceeds kT , has indicated an anomalous, non-thermalized population that cannot be described using the same Boltzmann temperature that characterizes the lower rotational levels (Cosby and Slinger, 2007; Dodd et al., 1994; Kalogerakis, 2019; Noll et al., 2015; Pendleton et al., 1989; Pendleton et al., 1993). In keeping with the terminology employed by Pendleton et al. (1993), these two conditions are referred to as Local Thermodynamic Equilibrium (LTE) and Non-Local Thermodynamic Equilibrium (NLTE), respectively. The term LTE as used here is not technically correct as it does not account for radiative effects. Instead it relates only to the collisional distribution of the rotational levels being characterized by the temperature of the surrounding gas, as has been done in Pendleton et al. (1993). Work is currently underway to use observations of the excess populations in the high rotational levels of the OH airglow to determine state-to-state quenching coefficients and to understand the thermalisation process in OH (Kalogerakis et al., 2018).”

P.2, L.8: There are additional relevant studies by Kalogerakis et al. published in ACP and Science Advances in 2019.

See new text in the previous comment.

P.2, L.10: "the OH is in LTE": This statement is wrong. As the authors use an OH kinetic model involving non-LTE chemistry and radiation to derive the OH layer depending on vibrational level, LTE is only true for the rotational populations of a fixed vibrational level in the model. In the case of full LTE, most OH molecules would be in the vibrational ground state. Also note that a difference in the effective emission altitude for the different vibrational levels is a strong indicator of non-LTE effects. Larger non-LTE excesses are related to higher emission altitudes.

In addition to the responses to P1, L32 and P2, L1 we have changed this phrase to "the rotational population distribution of the OH is in LTE" for clarification. The whole paragraph now reads:

"Here we examine the effects of temperature gradients in the OH emission region on the resulting vertically integrated spectrum of the Meinel $\Delta v=2$ sequence. To achieve this, model work was executed, where the model assumes that for each vibrational level, the rotational population distribution of the OH is in LTE at every altitude. The emission in each rotational line is then integrated vertically."

P.2, L.12-13: The use of "excess" is only correct if its definition is related to the lowest rotational levels. In the case of a temperature gradient of zero and an average temperature as the reference, there would also be negative population changes up to a certain rotational level.

We defined the word excess here as relative to the expected Boltzmann population due to a single temperature, which is characterised by the lines with $N \leq 4$. However, because these negative excess populations all are $<1\%$ and very small compared to the effects of higher lines, we decided to keep the term. We agree however that the term should be better defined here (see also comment below) and we have added a further sentence here. The context now reads:

"We find that even if the OH rotational levels are in strict LTE with the surrounding atmosphere, the temperature gradients through the OH emission region will create apparent excess emission in the higher OH rotational lines. Here we calculate the apparent excess population relative to the Boltzmann population expected using the temperature determined by the population of rotational levels with $N \leq 4$. This excess population can be incorrectly interpreted as due to NLTE effects, affecting the subsequent calculations of the thermalisation process."

P.2, L.30: "the OH" should be extended by "rotational level distributions" (see comment on P.2, L.10).

We have changed the text accordingly. The whole sentence now reads:

"Thus the OH rotational level distributions, even if thermalised at each altitude, will be described by different temperatures at each of those altitudes."

P.2, L.30-32: In fact, there is not a single OH layer if non-LTE effects contribute and OH ro-vibrational states are considered separately. Each OH line originates from a different altitude range (even if the upper vibrational level is kept fixed) depending on the non-LTE contribution. Hence, in reality, it will be complicated to separate the temperature gradient effects from the true non-LTE effects.

The referee is of course right in saying that non-LTE effects will make the situation more complex. The aim of this paper is to demonstrate that even without NLTE effects there is an important impact on the rotational distribution of the OH airglow. We agree that it will be complicated to separate the temperature gradient effects from the true non-LTE effects. This paper makes no attempt to explain the observed NLTE effects. However, even with NLTE effects, the sentence in question here would still be true: "Any instrument that integrates through the OH layer will therefore not see rotational line emission resulting from a single, average temperature, but from the whole span of temperatures present in the layer." We do not see that this statement is in conflict with the reviewer's comment, and prefer to keep the sentence as is, but thank the referee for the further explanation.

P.2, L.32-33: "the emission from high rotational lines that occurs in the warm regions": This statement should be softened since a negligible contribution of the cold regions to the high rotational states would require large temperature gradients. In this respect, it would be helpful if such a criterion could be quantified in the paper.

We have added a citation to the paper of Picard et al. (2004), who measured temperature changes of 60 between 85 and 95 km altitude. Additionally, the reported wave amplitudes in the mesopause region are consistent with temperature gradients up to this order. Since such large perturbations due to waves/tides are possible and observed, and even smaller gradients are shown in our results to have a significant impact, this sentence would appear to be accurate.

P.3, L.8: "OH is in LTE" (see comment on P.2, L.10).

See also answer to the comment P.2, L.10. We have also made the wording here more precise. The sentence now reads:

“We utilise a steady-state model of the OH VER, described below, to synthesise individual synthetic rotational spectra at 1 km intervals from 74 to 110 km. The model assumes that for each vibrational level, the rotational population distribution of the OH is in LTE (i.e. a single-temperature Boltzmann distribution) with the local temperature at each altitude.”

P.3, L.11: "excess population" (see comment on P.2, L.12-13).

See also answer to comment P2, L12-13. After already defining the term more clearly, we feel confident in keeping this text.

P.4, L.5-7: The use of hot nascent OH populations stringently leads to non-LTE level populations. Therefore, it should be made clear that these calculations are only used to derive the vertical OH emission distribution for each vibrational level (which neglects the spread of emission heights depending on the rotational level). The rotational level population distributions are arbitrarily set to LTE.

We have specified more clearly that the rotational population calculations are performed for each vibrational level. The text now reads:

“was also used to calculate the production rate of OH for each vibrational level, using the reaction rate coefficient (Sander et al., 2003). Due to the exothermicity of reaction 4, vibrational levels from $v'=6$ to 9 can be populated. The production of each vibrational level OH*(v') is calculated using the branching ratios from Sander et al. (2019).”*

P.4, L.8: The most frequent constituent of the Earth's atmosphere, N₂, is not considered for the collisional loss. Why?

The rate coefficient for OH quenching with N₂ is small, and Knutsen et al. (1996) were only able to provide an upper limit. Since the O₂/N₂ mixing ratio is virtually constant up to the turbopause, the O₂ would be 3 times more effective at quenching than the N₂. Thus, neglecting the N₂ would be well within the uncertainty of the O₂ rate coefficient and would not significantly affect the altitude distribution of the OH.

P.4, L.10: Why is the primitive "sudden death" approach used for all kinds of collisions? This is not state of the art. There are various examples of more sophisticated models in the literature. For example, the model by Adler-Golden (1997) as well as the fast Sharma et al. (2015) process for OH vibrational relaxation by atomic oxygen collisions had a big impact.

We thank the referee for pointing this out. The more sophisticated quenching models primarily change the relative vibrational distributions. However, the altitude profile of a single vibrational level is relatively unchanged, which is the primary consideration in this work. We have compared the vibrational distribution of single vibrational levels generated here with those of Adler-Golden (1997) and found them to be in good agreement. A stronger effect on the altitude profile is the profile of atomic oxygen, which is known from the SABER measurements to be highly variable. However, for MSIS atomic oxygen distributions, we are confident that our simple model reproduces the altitude distribution of a single vibrational level adequately for the purposes of this study. A small addendum was added to the paragraph mentioning the comparison with the Adler-Golden results.

However, a mistake was made in the description of this model. The sudden death criterion is used for the O quenching, while O₂ and CO₂ quench for steps of one vibrational unit. These are the major quenching channels for these species (Chalamala and Copeland, 1993; Dodd et al., 1991; Dyer et al., 1997; Knutsen et al., 1996; McDade and Llewellyn, 1987). We have changed the text here to correct this mistake. It now reads:

“Collisional loss for each OH(v') vibrational level was calculated for collisions with O, O₂ and CO₂ using the rate coefficients of Dodd et al. (1991), Knutsen et al. (1996), Dyer et al. (1997) and Chalamala and Copeland (1993). The model assumes quenching to the ground vibrational state, known as “sudden death” for the O, and stepwise quenching by one vibrational unit for the O₂ and the CO₂ (McDade and Llewellyn, 1987). The relative shape and peak height of the altitude profiles of the individual vibrational levels agree closely with those of the more sophisticated model of Adler-Golden (1997)”*

P.4, L.11: Radiation is also an important source of non-LTE effects, especially if it is not reabsorbed nearby as in the case of OH. This also contributes to the fact that the model in Sect. 2.1 is not able to provide OH in LTE. Hence, one should make clear that there is no consistent OH model that delivers thermalised populations. The investigation of the temperature gradient effect therefore requires the arbitrary manipulation of population distributions. I do not criticise this approach but it should be better communicated.

We now clarify the “misuse” of the terms LTE and non-LTE when we first introduce them in the body of the paper. Please see also the answer to the comment on P1, L32 – P2, L1.

“The term LTE as used here is not technically correct as it does not account for radiative effects. Instead it relates only to the collisional distribution of the rotational levels being characterized by the temperature of the surrounding gas, as has been done in Pendleton et al. (1993). “

P.4, L.11: "z" is not defined.

We have added the definition of z as the altitude. The text now reads:

“The total radiative loss from each vibrational level is given by $N_{\nu}(z) / \tau_{\nu}$, where $N_{\nu}(z)$ is the concentration of the hydroxyl ν' vibrational level at the altitude z and its lifetime, τ_{ν} , are calculated from Mies (1974).”

P.4, L.12: Mies (1974) does not give lifetimes. He provides Einstein-A coefficients. The set of coefficients is already quite old. Better data would be available.

We thank the referee for this comment. It is true that Mies calculated Einstein coefficients rather than lifetimes. We used these to calculate the lifetimes ourselves. This is now written more precisely in the paper. (See new text in the previous comment.) However, while the distribution of line strengths within the rotational levels are different with different authors, primarily due to vibration-rotation coupling, these differences tend to average out. This is because the differences are in different directions for the R- and P- branches, and virtually zero for the Q-Branch. Thus the lifetimes of the vibrational levels (the inverse of the sum of the individual line strengths) is relatively unchanged.

P.4, L.13: Mies (1974) does not introduce $\omega_{\nu\nu'}$.

Mies (1974) does, however, give all the important Einstein coefficients to calculate the branching ratio as the ratio of the sum of A-coefficients for one level divided by the total sum of all A-coefficients. We have specified this in the text, which now reads:

“The total VER of any ν' to ν'' vibrational transition is then given by $V_{\nu\nu''}(z) = \omega_{\nu\nu''} \cdot N_{\nu}(z) / \tau_{\nu}$, where $\omega_{\nu\nu''}$ is the vibrational branching ratio calculated from the Einstein coefficients from Mies (1974).”

P.4, L.26-29: Eq. 5 needs to be revised for several reasons. $N_{J'}$ can only be a relative population as an absolute population is already given by $N_{v'}$ if the descriptions from Sect. 2.1 still hold. A better solution would be to use $N_{v'J'}$ from Eq. 6. As $\omega_{v'v''}/\tau_{v'}$ corresponds to $A_{v'v''}$, the equation shows a product of Einstein-A coefficients, which looks wrong. It would be OK to replace both terms by $A_{v'v''J'J''}$. Moreover, $V_{v'v''J'J''}$ and $N_{J'}$ are also functions of z . Finally, level splitting by spinorbit coupling is not considered (symbol: i or F). This should be included as Fig. 2 proves that it was considered for the model.

We have changed the equation according to the suggestions of the referee. We have also now explained that we only look specifically at the 3/2 electronic subset and drop all further references to the F quantum number for readability. The new passage now reads:

$$V_{v'v''J'J''}(z) = \frac{\omega_{v'v''}}{\tau_{v'}} N_{v'J'}(z) \cdot A_{v',v'',J'J''}, \quad (2)$$

where $A_{J'J''}$ is the Einstein coefficient for the rotational transition and $N_{v'J'}$ is the population of the upper rotational level, J' . We concentrate in this paper on the 3/2 electronic subset of the OH airglow and drop the spin-orbit splitting quantum number F in all equations for readability."

P.4, L.29 - P.5, L.2: Eq. 6 also needs to be revised since the partition function $Q_{v'}(T_{\text{rot}})$ (Mies 1974) is missing. Otherwise the rotational level population distribution is not correctly normalised. It would be better to replace $E_{J'}$ by $E_{v'J'}$ as the level energy also depends on v' . Moreover, the electronic substates are not provided. Finally, the z dependences of $N_{v'J'}$ and $N_{v'}$ are not given.

We have changed the equation according to the suggestions from the referee. The section now reads:

$$N_{v'J'}(z) = \frac{N_{v'}(z)2(2J'+1)}{Q_{v'}(T(z))} \cdot \exp\left(\frac{-E_{v'J'}}{k \cdot T(z)}\right) \quad (3)$$

Where $N_{v'}$ is the total population of the v' vibrational level from the model, $E_{v'J'}$ is the energy of the v',J' vibration-rotation level, the factor $2(2J'+1)$ is the degeneracy of that level (including Λ -doubling) and $Q_{v'}(T(z))$ is the partition function (Herzberg, 1950). "

P.5, L.4: "the OH is in LTE" (see comment on P.2, L.10).

Please see the answer to that comment. Again, here we state that the model assumes that the OH is in LTE to show that a single-temperature Boltzmann population, at the local temperature of the atmosphere, is used to describe the rotational distribution of emission.

P.5, L.7-8: The statement related to the cool and warm regions should be weakened since a complete separation of the rotational level populations cannot be expected for typical temperature gradients.

We have amended the sentence since, as the referee says, the low lines are not only due to the cooler regions. The whole sentence now reads:

“This results in a net spectrum of low rotational lines whose intensities are enhanced relative to the mean temperature of the emission region when emitted from the cooler regions. Accompanying this are high rotational lines whose intensities are enhanced relative to the mean temperature of the emission region when emitted from the warm regions. Each of these is weighted by the VER of the vibrational transition at each altitude, $N_v(z) \cdot \omega_{v'v''} / \tau_{v'}$.”

P.5, L.11: "assuming LTE" should be extended by "for the rotational level populations".

We agree that this addition makes the text more accurate. The complete sentence now reads:

“As an example, the model described above was used to create a synthetic spectrum of the (7,4) rotational-vibrational band, assuming LTE for the rotational level populations at every altitude level, for mid-July, mid-latitude conditions”.

P.6, L.2: "ergs cm⁻³ s⁻¹" are quite old-fashioned units. Moreover, "s⁻¹" would be correct.

The missing minus sign has been added. We prefer to keep the units in ergs cm⁻³ s⁻¹ for direct comparison with the units system used with the SABER instrument.

P.6, L.7-8: The statement is only correct for the plotted phase of the wave. A phase shift of 180 deg would change the effective sign of the temperature gradient in the OH layer.

This is true. Later in the paper we model all phases to cover such cases. In this figure caption we only refer to the example given in the figure to highlight the general effect. We have changed the sentence to make this clearer and to avoid confusion. It now reads:

“With the example wave given here, the temperature variation increases the VER of the lower rotational lines at 95 km altitude, and of the higher rotational lines at 80 km.”

P.6, L.10-13: In the same way as Eqs. 5 and 6, Eq. 8 needs to be improved. I propose to write $I_{v'v''J'J''}$, $A_{v'v''J'J''}$, $E_{v'J'}$, and $E_{v''}$. This is further modified if the electronic substates are introduced (e.g. by adding i' or F'). Finally, $Q_{v'}(T_{rot})$ is also missing here.

We have updated the equation as suggested. It now reads:

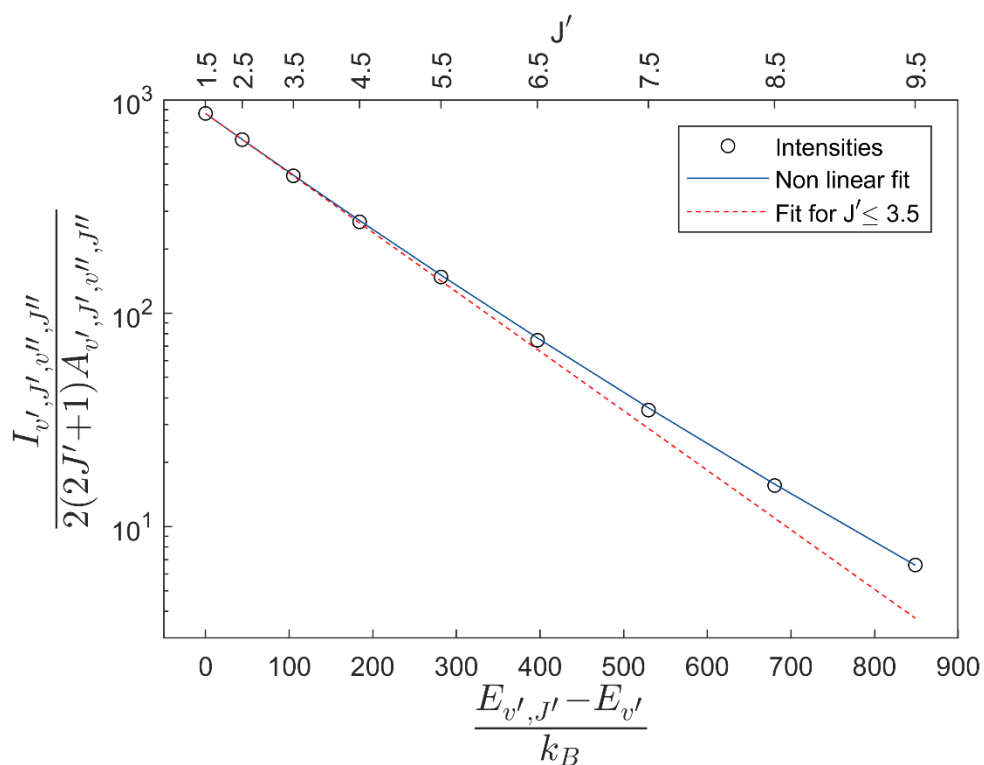
$$\frac{I_{v',v'',J',J''}}{2(2J'+1)A_{v',v'',J',J''}} = Q_{v'}(T)N_{v'} \cdot \exp\left(\frac{-(E_{v',J'}-E_{v'})}{kT}\right)$$

P.6, L.13-16: As Eq. 9 is very similar to Eq. 8, the same proposals are relevant. Depending on the changes, the symbols in Fig. 3 need to be adapted.

We have also updated this equation. It now reads:

$$\ln\left(\frac{I_{v',v'',J',J''}}{2(2J'+1)A_{v',v'',J',J''}}\right) = \ln(Q_{v'}(T)N_{v'}) - \frac{1}{T}\left[\frac{(E_{v',J'}-E_{v'})}{k}\right]$$

We have also updated the symbols in the Figure (which is now Figure 4):



P.7, L.1-2: "the lowest three rotational levels": This obviously refers to the $F = 1$ substates.

This is correct. Since we have now said specifically that we only look at the $F = 1$ sub-state (see comments to P4, L26-29), we keep the text here as it was.

P.7, L.10: The populations are related to P_1-branch lines.

Since we have now said specifically that we only look at the $F = 1$ substates (see comments above), we keep the text here as it was.

P.7, L.16 - P.8, L.1: This is again an equation which should be improved (see comments on previous equations).

We have updated this equation in the same manner. It now reads:

$$\ln \left(\frac{I_{v',v'',J',J''}}{2(2J'+1) \cdot A_{v',v'',J',J''}} \right) = \ln(Q_{v'}(T)N_{v'}) - \frac{1}{T} \left[\frac{(E_{v',J'} - E_{v'})}{k} \right] + \frac{\beta}{T^2} \left[\frac{(E_{v',J'} - E_{v'})}{k} \right]^2$$

P.8, L.1-2: It is stated that the beta parameter was used to improve the fit of the population distribution in Fig. 3. However, the actual value of beta is not provided.

The β factor for this example is 0.019. We have added this to the text. It now reads:

"The retrieved temperature $T_{NL} = 154.9 \pm 0.1$ K, is the same as $T_{1,3}$ within the fitting uncertainties, with a β factor of 0.019."

P.8, L.28-30: beta is also not given for the scenarios in Fig. 4.

The wave used for this figure is the same as used before, so the β -factor is still 0.019. We have added this information to the figure caption. It now reads:

"This is the same wave as presented in Figure 4. The distribution has a $\beta = 0.019$."

P.8, L.32-34: "the distribution of integrated rotational line intensities" for the fitting procedure does not appear to be limited in terms of the rotational levels. If high rotational levels contribute to the fit, the low rotational levels will show under populations. In this case, "excess" will be the wrong term.

The apparent excess population is defined relative to the predicted Boltzmann population of the lowest three rotational lines. This has also been clarified earlier (see comments above). We have also clarified this point in the text here. The text now reads:

"The apparent excess population is the ratio between the calculated intensity of a rotational line integrated in altitude, and the intensity of that line predicted by a Boltzmann distribution fitted to the distribution of integrated line intensities of the lowest three rotational lines using a single, effective temperature"

P.9, L.10: Excess populations for P₁-branch lines are shown. The excess is probably related to the first three rotational levels. This information should be given in the caption.

We agree that this should be stated here. The text now reads:

"The calculated apparent excess population of the OH (7,4) P(*J'*) lines relative to the Boltzmann population characterized by the temperature fitted to the lowest three lines."

P.9, L.14-26: The comparison with the measurements of Pendleton et al. (1993) would be less awkward if the population ratios from their Fig. 16 are also plotted in Fig. 4. It should not be a problem to add histograms of different colour or transparency to the two subfigures.

We agree that this is a sensible addition. We have changed the figure and associated text as follows:

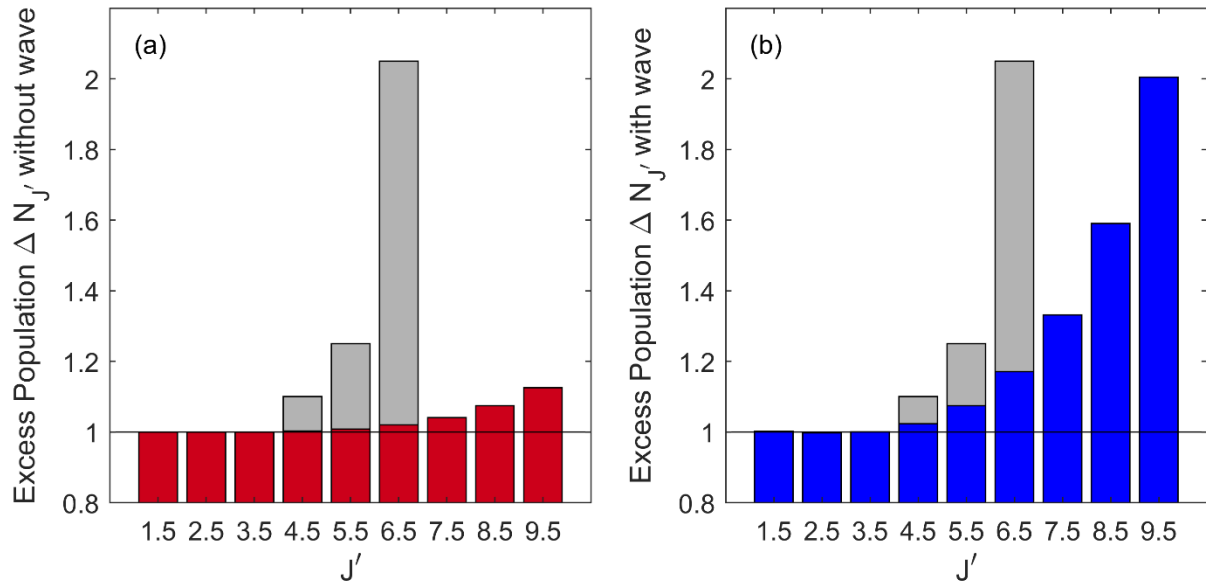


Figure 5: The calculated apparent excess population of the OH (7,4) P(J') lines relative to the Boltzmann population of the fitted temperature to the lowest three lines. Red on the left is calculated with the climatological temperature gradient shown from NRLMSISE-00 above Boulder, Colorado (40.0° N; 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).

These results can be compared to the findings from Pendleton et al. (1993) in their Fig. 16, for the (7,4) transition above Boulder, Colorado during mid-summer, the same season and transition as presented here. These results are shown as grey bars in Figure 5 (a) and (b)."

P.9, L.22: This is actually the only place where "P_1" is given. This notation will be understood if the related cases are consistently changed in the paper.

We have removed the 1 subscript, as we now discuss earlier in the paper that we are only discussing the 1 sublevel. The text now reads:

"For example, Pendleton et al. (1993) reported an apparent excess population of around a factor of 2 for the P($N=7$) (i.e. $J'=6.5$) line."

P.9, L.26 - P.10, L.2: This conclusion sounds more dramatic than it is since OH kinetic models are usually resolved in altitude, i.e. the models implicitly include the temperature gradient effect. Hence, comparisons with ground-based measurements should be trustworthy without any change in the modelling approach.

We agree models is the wrong word to use here. We are referring to calculations of kinetic thermalisation based on integrated airglow measurements. We have clarified this by changing the word "models" for "studies". The text now reads:

"Thus, the effect of the atmospheric temperature background has to be considered in addition to NLTE effects whenever the populations inferred from integrated airglow observations of high J' lines are to be used in kinetic thermalisation studies."

P.10, L.5: It is not clear which lines were used for the baseline fit for the derivation of the excess population. It might be helpful to define the term "excess population" at the first occasion and then to state that this definition is used for the rest of the paper.

We have defined "excess population" in response to the P2, L12-13 comment.

P.10, L.6-7: "The figure shows the (7,4) transition" sounds a bit strange. The figure shows results related to the (7,4) band. There are other similar sentences in the paper. As I would primarily expect that "transition" describes a spectral line, "band" or "vibrational transition" appear to be clearer.

We keep the nomenclature of Pendleton et al. (1993), who discuss the (7,4) "transition", as a comparison to our work. We agree however that this sentence is a bit clumsy and have rephrased it as suggested from the referee. It now reads:

"The figure shows results related to the (7,4) vibrational transition, which is the same transition that Pendleton et al. (1993) used."

P.10, L.9-10: The discussion in Sect. 3 only focusses on the "highest apparent excess population" for waves of a given amplitude and vertical wavelength. It would also be helpful to briefly quantify the average effect and the related variation. As already discussed in the context of the general comments, it is also important to know what the real distribution of temperature gradients for the OH layer is (or equivalently: the true frequency of the different simulated waves) in order to estimate the real impact of the temperature gradient effect. These results should be compared to the true non-LTE effects (as for the two examples in Fig. 4).

Such random wave motions can vary with latitude, longitude, season and time of day. We have attempted to give references in the paper indicating that the gradients observed here have been

observed. However, a comprehensive survey of all wave motions present in the mesosphere and their statistics is well beyond the scope of this paper.

However, we have added a short paragraph on the mean effect of waves with different phases as suggested. The mean effect is about 30 % of the maximum, which was presented in the paper, with a range between 20 % and 40 % dependent on the wavelength and amplitude of the wave. The new text is now:

“Figures 5 and 6 show the results for the phase which creates the strongest apparent excess population. The mean effect as a result of all different phases simulated is independent of the transition and varies between 20 and 40% of the maximum effect presented in Figures 5 and 6.”

P.12, L.3: Just writing "The apparent excess population" is incomplete. In fact, it is "The maximum apparent excess population" for the described wave.

We have changed the sentence as suggested. It now reads:

“The maximum apparent excess population of the $J' = 6.5$ upper level for a wave with wavelength 30 km and an amplitude of 30 K at 90 km altitude as a function of vibrational upper level.”

P.12, L.10: "the OH in LTE" (see comment on P.2, L.10).

See the response to that comment. Our simulations only calculate the OH in LTE, regardless of whether or not this happens in the real atmosphere. We changed the text slightly to make this even clearer:

“Even though the simulations calculated the rotational population distribution of the OH to be in LTE with the surrounding gas at every altitude, the integrated intensities of the higher rotational lines indicate an apparent excess population that could be misinterpreted as contributing to the NLTE effects previously reported (Cosby and Slinger, 2007; Noll et al., 2015; Pendleton et al., 1993).”

P.12, L.18: It depends on the research goal and the related significance of the effect whether the consideration of the effect is really "necessary". Concerning the estimate of the true impact, the paper should be improved.

We are confident that this paper shows that a considerable part of the ground-based measured excess population of higher rotational lines can be due to the influence of waves and tides. We therefore think it is important to consider this effect whenever rotational distributions are inferred

from ground-based airglow observations. This is exactly what the sentence in questions states. We have no intention to imply that these findings are “necessary” for anything other than this specific research goal. We are confident that the impact presented in this paper is a “true impact”, as the waves simulated in this paper have been observed before and could therefore be present in any ground-based measurement. Please see also the next comment.

P.12, L.18: "the rotational distribution of the OH" might be replaced by "OH rotational level population distributions", which sounds better.

We have changed the sentence as suggested. It now reads:

“We conclude that it is necessary to consider the temperature profile in order to infer OH rotational level population distributions from ground-based airglow observations.”

Technical corrections:

Whole paper: I have identified four different styles for the vibrational level symbol "v" (see e.g. P.5). In several equations, it is even an upper case letter. This should be harmonised. Other symbols should be checked in the same way.

We could not find the incident where the vibrational level was upper case. In equation (5), the capital V is referring to the volume emission rate rather than any vibrational level and should therefore be capitalised. We have attempted to harmonise the other instances as suggested by the referee.

P.9, L.2: "rad" should not be in italics.

We have changed this in the text.

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