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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Modelled Effects of Temperature Gradients and Waves on the Hydroxyl Rotational Distribution in Ground-Based Airglow Measurements" *by* Christoph Franzen et al.

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.

Specific comments:

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

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).

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).

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 thermalising 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.

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

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.

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.

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

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.

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.

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

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

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.

P.4, L.8: The most frequent constituent of the Earth's atmosphere, N_2, is not considered for the collisional loss. Why?

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.

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 is 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.

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

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.

P.4, L.13: Mies (1974) does not introduce omega_v'v".

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 spin-orbit coupling is not considered (symbol: i or F). This should be included as Fig. 2 proves that it was considered for the model.

P.4, L.29 - P.5, L.2: Eq. 6 also needs to be revised since the partition function Q_v'(T_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.

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

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.

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

P.6, L.2: "ergs cmˆ-3 sˆ1" are quite old-fashioned units. Moreover, "sˆ-1" would be correct.

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.

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.

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.

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

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

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

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.

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

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 underpopulations. In this case, "excess" will be a wrong term.

P.9, L.10: Excess populations for P_1-branch lines are shown. The excess is probably related to the first three rotational levels. This information should be given in the caption.

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.

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.

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.

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.

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.

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).

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.

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

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.

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

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.

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

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