

Review of the paper: "Hydroxyl airglow observations for investigating atmospheric dynamics: results and challenges" by Wüst et al. .

General comment.

This is excellent review paper. It can be accepted for publication in Atmospheric Chemistry and Physics after very minor correction which may take into account comments of other referees. I have just several minor comments.

Specific comments.

1. Introduction, 1<sup>st</sup> paragraph. Additionally were conducted number of rocket-borne measurements, for example MULTIFOT 92 (Takahashi et al., 1996).

2. Page 2. "Both parameters can vary (in the case of the centroid height by some kilometers) over several days or even during a single night due to strong dynamics (e.g., changes in the residual circulation during a stratospheric warming or strong tidal motions)". – Some modelling results show monthly averaged variation from ~78 km to ~91 km and in frame of single month it can be even stronger, specifically at high latitudes (Grygalashvily et al., 2014). This is much complex question than briefly noted here, because altitude variation of OH\* layer depends on latitude, season, vibrational number. From the other hand this is just introduction and should not cover all problems. Hence, authors may slightly extend this discussion or not, on their choice.

3. Page 2. "adjacent vibrational levels are separated by some 100 m (Baker and Stair, 1988; Adler-Golden, 1997)" it seems to me that a little bit stronger.

Backer and Stair (1988) found ~ 500 m, Adler-Golden (1997) found ~700 m, Grygalashvily et al. (2014) found 250-1000 m depending on season and latitude.

4. Page 3. The abbreviation MORTI should be disclosed.

5. Page 5, Section 2.1. Reaction 2 was proposed as hypothesis, which has not been confirmed and currently it is not considered.

6. Page 5, Section 2.1. Reaction 3. The reaction  $O+HO_2 \rightarrow OH^*+O_2$  was introduced as a source of vibrationally excited hydroxyl in the 1970s by, probably (?), Nagy et al. (1976) as a hypothesis put forward for energy reasons (the energy of this exothermic reaction is sufficient to produce  $OH^*$  up to and including the 6<sup>th</sup> vibrational level) and was applied by several authors in the 1980s to explain discrepancies between observed emissions and calculation results (Takahashi and Batista, 1981; Turnbull and Lowe, 1983). At that time there were no sufficiently good measurements and calculations of molecular and atomic oxygen quenching coefficients, spontaneous emission coefficients and yield coefficients of the reaction of ozone with atomic hydrogen. But already Llewellyn et al. (1978) noted that with the new quenching coefficients they calculated, a new  $OH^*$  source (R3) would no longer be necessary. Further, Kaye (1988) showed from laboratory measurements that population above the 3<sup>rd</sup> vibrational level is not possible. Moreover, population coefficients for the first three levels have been proposed (Makhlouf et al., 1995) using general considerations without solid confirmations. To date, no more precise information on the exit coefficients has been obtained. Furthermore, with new calculated and laboratory-derived quenching coefficients, spontaneous emission coefficients, and yield coefficients for the ozone and hydrogen atom reaction, the application of hydroperoxide and oxygen atom reaction to obtain agreement on  $OH^*$  emission measurements is not required (Xu et al., 2012; McDade and Llewellyn, 1987). Although some authors still apply this reaction, it can be omitted from consideration until the time when it will be supported based on laboratory measurements.

If discussion in Section 2.1. cover day and night conditions, may be for authors will be interesting that recently was shown that water vapour dissociation may essential contribute to daytime  $OH^*$  population.

Chang, Y., Li, Q. M., An, F., Luo, Z. J., Zhao, Y., Yu, Y., He, Z., Chen, Z., Che, L., Ding, H., Zhang, W., Wu, G., Hu, X., Xie, D., Plane, J. M. C., Feng, W., Western, C. M., Ashfold, M. N. R., Yuan, K., & Yang, X. (2020). Water photolysis and its contributions to the hydroxyl dayglow emissions in the atmospheres of Earth and Mars. *Journal of Physical Chemistry Letters*, 11, 9086-9092. <https://doi.org/10.1021/acs.jpcllett.0c02803>

7. Page 5, Section 2.1. “Lower levels are populated in a radiative cascade by spontaneous emission” – I would not use the word “cascade” because it is related to “cascade” scheme at which the excited molecule relaxes to the one vibrationally excited level below (e.g. McDade

and Llewellyn, 1987), but in nature all quenching and spontaneous emission processes are neither “cascade” nor “sudden death”, but of multi-quantum relaxation nature.

8. Figure 1b, left panel. Why the nighttime atomic oxygen concentration, taking into an account discussion above, is higher than that for daytime at ~80-85 km?

9. Figure 1b, right panel. Currently well known that OH\*-layers with higher vibrational numbers have peaks higher than those for smaller vibrational numbers (e.g., McDade, 1991; Adler-Golden, 1997; and references therein). On this figure the sequence of vibrational numbers from higher altitude downwards is as follow: 5-4-7-6-3-2.

Why it is?

The number density of OH $\nu$  at peak grows in the direction of the smaller  $\nu$  (e.g., Sivjee and Hamwey, 1987; McDade, 1991; Adler-Golden, 1997; Xu et al., 2012; Caridade et al., 2013) On this figure the sequence of vibration numbers from lower concentrations toward higher is as follow: 6-5-4-7-3-2.

Why it is?

On my opinion this is not the best illustration.

All of the above are non-binding corrections to the article and are left to the authors' discretion. All references mentioned in the review are not required to be cited in the article.

Generally, after specific and technical corrections, I recommend this paper for publication in Atmospheric Chemistry and Physics.