



Several notes on the OH* layer

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Received: 7 June 2015 – Accepted: 15 July 2015 – Published: 27 July 2015

Abstract. This brief note introduces several analytical approaches to OH* layer parameters. The number density and height of the OH* layer peak are determined by the distributions of atomic oxygen and temperature, and by corresponding vertical gradients. The theory can be applied to satellite-borne and ground-based airglow measurements, as well as to model results.

Keywords. Atmospheric composition and structure (airglow and aurora; middle atmosphere – composition and chemistry; thermosphere-composition and chemistry)

1 Introduction

The field of applications of airglow has essentially large borders. It is widely utilized for temperature measurements in the mesopause region (e.g., Offermann and Gerndt, 1990; She and Lowe, 1998; Innis et al., 2001; Bittner et al., 2000, 2002; Espy et al., 1995, 2007). A large number of observations are directed at dynamic processes, e.g., gravity waves (GWs), planetary waves, and tides (Tarasick and Shepherd, 1992; Taylor et al., 1991, 1995a, b, 1997; Makhlof et al., 1995, 1998; Offermann et al., 2009, Shepherd et al., 2012, and references therein). Minor chemical constituents in the mesopause, which are scarcely retrieved by other means, e.g., atomic oxygen (Russell et al., 2005; Smith et al., 2010), ozone (Smith et al., 2008, 2009), and atomic hydrogen (Thomas, 1990; Takahashi et al., 1996; Mlynczak et al., 2014), were measured by the observation of airglow emission. One of the subjects of investigations by airglow measurements is the trend of temperature (e.g., Espy and Stegman, 2002; Offermann et al., 2006, 2010). Potentially, airglow measurements can be useful to retrieve water vapor in the mesopause (Kulikov et al., 2009) and chemical heat (Mlynczak and Solomon, 1993).

Despite large application airglow measurements, hydroxyl layer morphology has yet to be studied intensively. The still hidden area of knowledge is information about layer altitude variability with the diurnal cycle, in the context of seasonal-latitudinal cycles, due to GWs, etc., and questions on correlations between altitude of the layer, number density (volume emission, intensity), winds (meridional and vertical), etc. The same questions are related to long-term changes in the OH* layer. Unfortunately, satellite airglow measurements have been applied only recently and may not have the ability to offer conclusions about trends of the OH* layer. While ground-based measurements are limited by local latitude and integrated volume emission, the retrieval of latitudinal variation and emission peak are difficult.

Notwithstanding large applications and some amount of empirical information, there is a lack of analytical understanding of OH* layer behavior. Most part of theoretical investigations with OH* layer has focused on numerical simulations. In order to provide a somewhat theoretical approximation, we utilize parameters that describe morphology and variability of the layer. The natural choice for such parameters is number density at peak and altitude of peak. In the next chapter, we present some theoretical approximations of altitude and number density at the peak of the OH* layer.

2 Theory

We assume the photochemical equilibrium for excited hydroxyl in the mesopause region; hence, we can write this as the ratio of production term P_v to the loss term L_v :

$$[\text{OH}_v] = \frac{P_v}{L_v}. \quad (1)$$

Hereafter, the square brackets, as traditionally, denote the number density of given constituents. In the production

and loss terms, contributions from the chemical reactions, quenching by O₂, N₂, and O, and spontaneous emission are summarized. The most general form for the production term of OH_v* is

$$\begin{aligned}
 P_v &= k_1(v) [\text{O}_3] [\text{H}] + k_2(v) [\text{HO}_2] [\text{O}] \\
 &+ \sum_{v'=v+1}^9 q_{v'v} [\text{OH}_{v'}] [\text{O}] \\
 &+ \sum_{v'=v+1}^9 \tilde{q}_{v'v} [\text{OH}_{v'}] [\text{N}_2] + \sum_{v'=v+1}^9 Q_{vv'} [\text{OH}_{v'}] [\text{O}_2] \\
 &+ \sum_{v'=v+1}^9 A_{v'v} [\text{OH}_{v'}], \quad (v' > v), \quad (2)
 \end{aligned}$$

where k_i represents the reaction rates; v is the vibrational number; q , \tilde{q} , Q are the rates for quenching by atomic oxygen, molecular nitrogen, and molecular oxygen, respectively; and $A_{v'v}$ are the Einstein coefficients for spontaneous emission (e.g., Turnbull and Lowe, 1989). The first term of Eq. (2) corresponds to the main source of the excited hydroxyl in the mesopause ($\text{H} + \text{O}_3 \rightarrow \text{OH}_{v=1-9}^* + \text{O}_2$), which is responsible for the population of the first nine vibrationally excited levels (Bates and Nicolet, 1950). The second term is the minor source of excited hydroxyl in the mesopause region: $\text{O} + \text{HO}_2 \rightarrow \text{OH}_{v=1-6}^* + \text{O}_2$ (Breig, 1970; Nagy et al., 1976), which populates no higher than six vibrational levels and is possibly important at high latitudes (e.g., Takahashi and Batista, 1981; Lopez-Moreno et al., 1987; Kaye, 1988). The third, fourth, and fifth terms represent transitions from the highest vibrational levels due to quenching by atomic oxygen, molecular nitrogen, and molecular oxygen, respectively. The spontaneous emission is represented by the last term.

The most general loss term,

$$\begin{aligned}
 L_v &= k_3(v) [\text{O}] + \sum_{v''=0}^{v-1} q_{vv''} [\text{O}] + \sum_{v''=0}^{v-1} \tilde{q}_{vv''} [\text{N}_2] \\
 &+ \sum_{v''=0}^{v-1} Q_{vv''} [\text{O}_2] + \sum_{v''=0}^{v-1} A_{vv''}, \quad (v'' < v), \quad (3)
 \end{aligned}$$

among the previously mentioned processes, takes into an account the chemical removal (first term) by the reaction $\text{OH}^* + \text{O} \rightarrow \text{H} + \text{O}_2$ with the reaction rate $k_3(v)$ (Varandas et al., 2004). The joint system of Eqs. (1), (2), and (3) is extremely complex and nonlinear; thus, for analysis, it should be simplified.

2.1 Zero approximation: “sudden death” quenching by O₂

We are searching for a simpler solution in the region of the OH* layer and at nighttime conditions. In this region, the production of OH* is primary governed by the reaction $\text{H} +$

$\text{O}_3 \rightarrow \text{OH}_{v=1-9}^* + \text{O}_2$, thus the production term

$$P_{\text{OH}^*} \approx k_1(v) [\text{H}] [\text{O}_3] = r_1 \times f_v [\text{H}] [\text{O}_3], \quad (4)$$

where the reaction rate $r_1 = 1.4 \times 10^{-10} \exp(-460/T)$ (Sander et al., 2011), and f_v is the nascent distribution.

In the view of ozone balance, assuming ozone in the photochemical equilibrium ($d[\text{O}_3]/dt = 0$) (Smith et al., 2008) equating production and loss of the ozone at nighttime conditions, we can write

$$r_4 [\text{O}] [\text{O}_2] [M] = r_1 [\text{H}] [\text{O}_3] + r_5 [\text{O}] [\text{O}_3], \quad (5)$$

where M is the air number density, $r_4 = 6 \times 10^{-34} (300/T)^{2.4}$ and $r_5 = 8 \times 10^{-12} \exp(-2060/T)$ are the reaction rates for $\text{O} + \text{O}_2 + M \rightarrow \text{O}_3 + M$ and $\text{O} + \text{O}_3 \rightarrow 2\text{O}_2$, respectively. The share of the reaction of ozone with atomic oxygen in total ozone loss is no larger than 8% in the region of OH* peak (Smith et al., 2008), since r_5 is several orders smaller than r_4 in this region. Thus, the second term in the right-hand side of Eq. (5) can be omitted. Substituting Eq. (5) into Eq. (4), we obtain the following:

$$P_{\text{OH}^*} \approx f_v r_4 [\text{O}] [\text{O}_2] [M]. \quad (6)$$

The quenching by molecular oxygen is assumed to be a main loss process (Adler-Golden, 1997; Knutsen et al., 1996). The most simple parameterization for quenching processes is “sudden death”, when relaxation is assumed to be due simply to transitions from the given vibrational level to zero (Llewellyn et al., 1978; McDade and Llewellyn, 1987). In this case, we can write the loss term Eq. (3) as follows:

$$L_v \approx \hat{Q}_v [\text{O}_2], \quad (7)$$

where \hat{Q}_v is “sudden death” quenching rates for molecular oxygen.

Hence, from Eqs. (1), (7), and (6), with explicitly written r_4

$$[\text{OH}_v] \approx \frac{f_v \times 6 \times 10^{-34} (300/T)^{2.4} [\text{O}] [\text{O}_2] [M]}{\hat{Q}_v [\text{O}_2]}. \quad (8)$$

With the ideal gas law ($[M] = p/k_b T$, where k_b is the Boltzmann constant) and omitting $[\text{O}_2]$:

$$[\text{OH}_v] \approx \frac{f_v 6 \times 10^{-34} \times 300^{2.4}}{k_b \hat{Q}_v} \times p T^{-3.4} [\text{O}] = B \hat{C}_v p T^{-3.4} [\text{O}], \quad (9)$$

where $\hat{C}_v = f_v/\hat{Q}_v$ is the vibrationally dependent constant, and $B = 6 \times 10^{-34} \times 300^{2.4}/k_b$ is the vibrationally independent constant.

Differentiating Eq. (9) by p and equating it to zero, we obtain

$$\frac{\partial [\text{OH}_v]}{\partial p} = B \hat{C}_v \left(T^{-3.4} [\text{O}] + p \left(T^{-3.4} \frac{\partial [\text{O}]}{\partial p} - 3.4 T^{-4.4} [\text{O}] \frac{\partial T}{\partial p} \right) \right) = 0. \quad (10)$$

From Eq. (10), the altitude of OH* peak in pressure coordinates is

$$p_{\text{peak}} \approx \frac{[\text{O}]}{3.4 \times T^{-1} [\text{O}] \frac{\partial T}{\partial p} - \frac{\partial [\text{O}]}{\partial p}} = \frac{1}{3.4 \frac{\partial \ln T}{\partial p} - \frac{\partial \ln [\text{O}]}{\partial p}} = \frac{1}{\frac{\partial(3.4 \ln T - \ln [\text{O}])}{\partial p}} = \frac{1}{\frac{\partial}{\partial p} \left(\ln \left(\frac{T^{3.4}}{[\text{O}]} \right) \right)} \quad (11)$$

From Eq. (11) in Eq. (9), the number density at the peak of the OH* layer is

$$[\text{OH}_v]_{\text{peak}} \approx \frac{B \widehat{C}_v T^{-3.4} [\text{O}]}{\frac{\partial}{\partial p} \left(\ln \left(\frac{T^{3.4}}{[\text{O}]} \right) \right)} \quad (12)$$

Already in the context of this simplest approximation, we can explain large numbers of empirical phenomena related to the OH* layer, for example, height-number density anticorrelation. However, well-known vertical separation with respect to vibrational numbers has not been explained because Eq. (11) does not depend on v . Moreover, all explanations regarding this approximation are just qualitative because the simplest assumption of “sudden death” quenching by O₂ does not give a correct quantitative assessment.

2.2 First approximation: multi-quantum quenching by O₂

Most natural case for quenching processes is multi-quantum relaxation when taking into an account transitions from all the highest to all the lowest vibrational levels. In this case, the ratio of the production to the loss term is as follows:

$$[\text{OH}_v] = \frac{r_1 f_v [\text{O}_3] [\text{H}] + \sum_{v'=v+1}^9 Q_{v'v} [\text{OH}_{v'}] [\text{O}_2]}{\sum_{v''=0}^{v-1} Q_{vv''} [\text{O}_2]} \quad (13)$$

where Q – multi-quantum quenching rates for molecular oxygen.

Taking into an account all assumptions as before and reducing [O₂], Eq. (13) can be transformed:

$$[\text{OH}_v] = \frac{f_v B T^{-3.4} [\text{O}] p + \sum_{v'=v+1}^9 Q_{v'v} [\text{OH}_{v'}]}{\sum_{v''=0}^{v-1} Q_{vv''}} \quad (14)$$

Writing consequently [OH_v] for $v = 9, \dots, 1$, we can infer identical to the Eq. (9) shape of expression for the exited hydroxyl:

$$[\text{OH}_v] \approx B \widetilde{C}_v p T^{-3.4} [\text{O}], \quad (15)$$

but with the new, vibrationally dependent coefficient \widetilde{C}_v , which has a recursive character, it is now

$$\widetilde{C}_v = \frac{f_v + \sum_{v'=v+1}^9 \widetilde{C}_{v'} Q_{v'v}}{\sum_{v''=0}^{v-1} Q_{vv''}} \quad (16)$$

Differentiating Eq. (15), as before, by p and equating it to zero, we obtain a solution identical to the Eq. (11) solution for OH* peak, but now the concentrations at the peak of the layer are more realistic according to the more realistic quenching process. Nevertheless, layers with different vibrational numbers are still not separated in altitude.

2.3 Second approximation: “sudden death” quenching by O and O₂

In the simplest case of “sudden death” quenching by O₂, we add a second important quencher – atomic oxygen with the same “sudden death” approximation and assumptions as before:

$$[\text{OH}_v] \approx \frac{r_1 f_v [\text{O}_3] [\text{H}]}{\widehat{Q}_v [\text{O}_2] + \widehat{q}_v [\text{O}]} \approx \frac{f_v \times B [\text{O}] [\text{O}_2] p T^{-3.4}}{\widehat{Q}_v [\text{O}_2] + \widehat{q}_v [\text{O}]}, \quad (17)$$

where \widehat{q}_v is the “sudden death” quenching rate for atomic oxygen.

As usual, examining Eq. (17) for the extremum with respect to pressure, we obtain

$$p_{v-\text{peak}} \approx \frac{1}{\frac{\partial}{\partial p} \left(\ln \left(\frac{(\widehat{Q}_v [\text{O}_2] + \widehat{q}_v [\text{O}]) T^{3.4}}{[\text{O}] [\text{O}_2]} \right) \right)} \quad (18)$$

Note, that now [OH_v] peak altitudes depend on vibrational numbers. Thus, quenching by atomic oxygen is responsible for the vertical separation of [OH_v] with a different v .

Substituting Eq. (18) in Eq. (17), we obtain an expression for OH* layer peak as the following:

$$[\text{OH}_v]_{\text{peak}} \approx \frac{f_v \times B [\text{O}] [\text{O}_2] T^{-3.4}}{\widehat{Q}_v [\text{O}_2] + \widehat{q}_v [\text{O}]} \times p_{v-\text{peak}} \quad (19)$$

Alternatively, taking into account the linear proportionality of molecular oxygen to the number density [O₂] = $\alpha [M]$ in OH* layer region (80–95 km, where $\alpha \approx 0.2$), Eq. (17) can be written as follows:

$$[\text{OH}_v] \approx \frac{f_v \times B \times B_2 [\text{O}] p^2 T^{-4.4}}{B_2 \widehat{Q}_v p T^{-1} + \widehat{q}_v [\text{O}]}, \quad (20)$$

where $B_2 = \alpha / k_b$.

Differentiating Eq. (20) by p and equating it to zero, we get cubic equation with $p_0 = 0$, and two other solutions are

quadratic with respect to the p equation:

$$\begin{aligned}
 ap^2 + bp + c &= 0, \\
 a &= \widehat{Q}_v B_2 \frac{\partial}{\partial p} \left(\ln \frac{[\text{O}]}{T^{3.4}} \right), \\
 b &= \widehat{Q}_v B_2 - 4.4 \widehat{q}_v [\text{O}] \frac{\partial T}{\partial p}, \\
 c &= 2 \widehat{q}_v [\text{O}] T,
 \end{aligned} \tag{21}$$

where a , b , and c are the leading coefficient, second coefficient, and free term, respectively.

3 Discussion

Annual variations of OH* layer altitude and number density were observed by SABER (Sounding of the Atmosphere using Broadband Emission Radiometry) and WINDII (Wind Imaging Interferometer) satellite measurements, simulated by TIME-GCM (Thermosphere–Ionosphere–Mesosphere Electrodynamics General Circulation Model) and ROSE model (Marsh et al., 2006; Liu et al., 2008; Gao et al., 2010). At high and middle latitudes, the lowest altitudes and largest number densities of the layer were found in winter and the highest altitudes with smaller number densities in summer. In the works mentioned above, the annual variability of general mean circulation and corresponding atomic oxygen transport were found to be a reason for annual cycle of OH* layer. From Eq. (9), we can see that OH* number density (and, consequently, intensity) is directly proportional to the atomic oxygen concentration. In Eq. (11), pressure increases with the growth of atomic oxygen concentration; consequently, the height of the layer declines and vice versa. The downward component of the residual circulation delivers atomic oxygen in the region of OH* layer in winter, and the upward component reduces atomic oxygen in summer. Thus, already when considering zero approximation, we can qualitatively explain the annual cycles of the OH* layer.

The middle and equatorial latitudes are characterized by semi-annual variations with maxima number density around the equinoxes and corresponding minima of the layer altitude (Wiens and Weill, 1973; Fukuyama, 1977; Marsh et al., 2006; Liu et al., 2008; Gao et al., 2010; Xu et al., 2010). The semi-annual variability of OH* was related to the semi-annual behavior of tides near the equator. The alternative point of view is that the semi-annual variation of OH* at low latitudes is related to seasonal variability of GWs and changes in turbulent diffusion due to GW dissipation (Garcia and Solomon, 1985). In view of new knowledge on GW mixing (Grygalashvily et al., 2011, 2012), it can be stated that the semi-annual variation of OH* is related to down-gradient fluxes of atomic oxygen by GW mixing. All three potentially responsible processes are connected to the downward flux of atomic oxygen, which, according to Eqs. (9) and (11), de-

termines number density and peak altitude of the OH* layer, respectively.

Given annual and semi-annual variability, one can infer an anti-correlation of height and number density, which is proportional to the intensity and volume emission (e.g., Baker et al., 2007; Gao et al., 2010). This subject was empirically investigated in a number of works (Yee et al., 1997; Liu and Shepherd, 2006; Mulligan et al., 2009). Liu and Shepherd (2006), based on WINDII measurements for the latitude band 40° S–40° N and the period from November 1991 to August 1997, derived an empirical formula for the dependence of the height of the layer on integrated intensity (that is equivalent to number density), day of year, and local time. Then, they subdivided latitudinal band into five bins and derived coefficients for each one. In a later study, the coefficients for the given empirical formula were derived for 78 ± 5° N (Mulligan et al., 2009). This is evident from analytical Eq. (9), which shows that the number density (and, consequently, the intensity and volume emission rate) of the OH* layer is directly proportional to pressure and, thus, inversely proportional to altitude.

The terannual variability of the OH* layer observed by ground-based methods (Fukuyama, 1977) and recently confirmed by modeling (Sonnemann et al., 2015) can be explained as the superposition of annual and semi-annual cycles with phase shift and amplitude maxima at different latitudes.

Only one work is currently devoted to relationships between OH* peak altitude and meridional wind flow (Dyrland et al., 2010). In this research, an anti-correlation between OH* peak altitude and meridional wind flow at high latitude (78° N) in winter (25 October–11 February) was found. Considering Eq. (11), which shows the inverse relationship between OH* peak altitude and atomic oxygen, taking into an account that atomic oxygen in this region has a negative meridional gradient (Smith et al., 2010), the anti-correlation between positive meridional wind flow and OH* layer altitude (at high latitudes in winter) due to the poleward transport of atomic oxygen becomes obvious.

The impact of sudden stratospheric warming (SSW) on the OH* layer has been studied intensively in several recent works (Dyrland et al., 2010; Shepherd et al., 2010; Damiani et al., 2010; Gao et al., 2011). During SSW, the altitude of the layer rises by ~5–7 km, and the number density is reduced by more than 50%. The reasons for such a reduction, as shown in the studies cited above, are the poleward meridional and upward vertical fluxes, which are typical for SSW events. Such a response of the OH* layer on SSW agrees with the notion about direct proportionality of excited hydroxyl number density to atomic oxygen (Eqs. 11 and 12).

Airglow intensity correlates with the Lyman- α 11-year solar cycle (Fukuyama, 1977; Pertsev and Perminov, 2008; Wang et al., 2013; Sonnemann et al., 2015). Indeed, years of solar maxima are characterized by stronger dissociation of molecular oxygen by the Lyman- α radiation at the

mesopause/lower thermosphere and, consequently, enhanced production of atomic oxygen. Aligning with Eq. (9), the number density of the OH* layer (hence, volume emission and intensity of airglow) correlates with the Lyman- α flux.

In the context of multi-quantum quenching by molecular oxygen, we obtain correct distributions of number densities using vibrational numbers. In Eq. (16), one can see that the numerator increases with decreasing v , while the denominator decreases. Hence, the number density Eq. (15) is proportional to the vibrationally dependent coefficient of Eq. (16), growing in the direction of the smaller v , as is well known from earlier results of modeling and measurements (e.g., Sivjee and Hamwey, 1987; Lopez-Moreno et al., 1987; McDade, 1991; Adler-Golden, 1997; Xu et al., 2012; Caridade et al., 2013).

Only with the consideration of atomic oxygen as a second quencher do we obtain vertical separation using vibrational numbers in Eq. (18). Since the works by Adler-Golden (1997) and Swenson and Gardner (1998), it has been well known that the altitude difference between the OH* layer peaks with different vibrational numbers depends on quenching by atomic oxygen. That is to say, more reactive quenching or larger values of atomic oxygen concentration are linked to stronger altitude differences between OH* peaks with different vibrational numbers. Obviously, in the absence of quenching by atomic oxygen, the terms with \hat{q}_v becoming zero, and molecular oxygen reducing, Eqs. (17), (18), and (19) become identical to Eqs. (9), (11), and (12), respectively. Thus, with reduced quenching by the atomic oxygen, the system of vertically separated peaks tends to the case with all peaks at the same altitude, and the distance between peaks with different v decreases.

Fichtelmann and Sonnemann (1987), based on an extremely simplified zero-dimensional chemical model, first showed that an enforced nonlinear photochemical system of the mesopause region manifested the day-to-day variation, which is controlled by strange attractor (particularly in O₃-OH phase-space), and a chaotic regime, analogous to other nonlinear systems. The nonlinear resonance can be evidenced if the eigenfrequency of homogeneous equations (characteristic time of photochemical system) is equal to the external frequency of stimulation (Sonnemann and Fichtelmann, 1987). Later on, Yang and Brasseur (1994) demonstrated that the photochemical system of the mesopause region has a trigger solution under conditions associated with a hydrogen flux, and, apparently, the system tends to create bi-stable behavior in that domain. The availability of two solutions of Eq. (21) points to the possible bi-stable behavior of the OH* layer peak. Realistic modeling at different parameters (e.g., water vapor, vertical eddy diffusion coefficient) related to the mesopause conditions infers period 2 oscillations that trigger 2-day variations in odd-oxygen/odd-hydrogen concentrations (Fichtelmann and Sonnemann, 1992; Sonnemann and Fichtelmann, 1997; Sonnemann and Feigin, 1999a, b; Sonnemann et al., 1999). Often, airglow measure-

ments infer 2-day oscillations in the temperature and wind data in the mesopause region (e.g., Ward et al., 1996; Takahashi et al., 2005; López-González et al., 2009; Pedatella and Forbes, 2012, and references therein). Bi-stable behavior of the OH* peak altitude and number density may result in a 2-day signal registered by measurements.

4 Conclusions

The expressions that determine the altitude at peak and number density at peak of the OH* layer were derived from several different approximations. At all approximations, OH* number density in the vicinity of the OH* layer is directly proportional to the atomic oxygen concentration and inversely proportional to the power of temperature. Given all approximations, the peak of the layer number density is anti-correlated with the height of the peak. Atomic oxygen is responsible for the vertical separation of sub-layers with different vibrational numbers and for the distance between them. The distance is proportional to the atomic oxygen number density. These approximations are useful for the interpretation of several empirical phenomena.

Acknowledgements. The author gratefully acknowledges the useful remarks and helpful discussions with G. R. Sonnemann and E. Becker.

The topical editor C. Jacobi thanks one anonymous referee for help in evaluating this paper.

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