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Note on consistency between Kalogerakis– Sharma Mechanism (KSM) and two-step mechanism of atmospheric band emission (762 nm)

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Abstract

For more than 30 years, a two-step mechanism was used to explain observed Atmospheric band emission (762 nm) in mesopause region. A new mechanism, which leads to the formation of electronically excited molecular oxygen that gives this emission, was proposed recently. We show, based on an analytical solution, that the fit-functions for Atmospheric band volume emission in the case of the two-step mechanism and the new Kalogerakis–Sharma Mechanism (KSM) have analogous expression. This derivation solves the problem of consistency between the well-known two-step mechanism and the newly proposed KSM.

Keywords: Atmospheric band emission, Kalogerakis–Sharma Mechanism, Airglow, Two-step mechanism, Mesopause region

Introduction

Airglow phenomena, and particularly atmospheric band emission (762 nm), attract interest, because they help infer knowledge about dynamical variabilities [e.g. tides, planetary waves (PWs), gravity waves (GWs)], temperature, and chemical distributions in the mesopause region. Atmospheric band emission (762 nm) is produced by the excited state of molecular oxygen $O_2\left(b^1\Sigma_g^+,\nu=0\right)$ (hereafter, ν is the vibrational number). It was used to investigate parameters of GWs (e.g. Noxon 1978; Zhang et al. 1993; Leko et al. 2002). PWs were studied by Lopez-Gonzalez et al. (2009). The tides have been detected, for example, by Lopez-Gonzalez et al. (2010, 2011) utilised this emission to infer the temperature in the mesopause region.

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Distributions of minor chemical constituents (ozone, atomic oxygen) were measured in a number of works (e.g. Mlynczak et al. 2001; Hedin et al. 2009, and references therein). The same excited state of molecular oxygen $O_2(b^1\Sigma_g^+)$ produces emission at 865 nm, which can be measured from the ground, and which has been utilised to study solar cycle effects (e.g. Pertsev and Perminov 2008), trends (e.g. Dalin et al. 2020, and references therein), and GWs (Aushev et al. 2000). This emission was utilised to investigate variability due to sudden stratospheric warming (Shepherd et al. 2010). The processes of energy transfer between different excited states of molecules and atoms, as well as the parameters of transitions, should be well studied for three main reasons: for correct interpretation of the results of measurements, for correct assessment thermalisation of upcoming radiation (that is significant for dynamics), and because the chemical properties of electronically excited states of atoms and molecules are different from their ground states (distributions of excited states are important for mesopause chemistry).



For over 30 years, a two-step (Barth-like) mechanism (Greer et al. 1981; Witt et al. 1984; McDade et al. 1986) was considered as the main source of $O_2(b^1\Sigma_g^+, \nu=0)$ population at nighttime conditions. It assumes production $O_2(b^1\Sigma_g^+, \nu=0)$ from the recombination of atomic oxygen via unknown precursor. The population ${\rm O}_2\!\left(b^1\Sigma_g^+, \nu=0\right)$ at daytime occurs by quenching of O(¹D), which is the result of ozone and molecular oxygen dissociation. (e.g. Yankovsky and Vorobeva 2020). It was assumed until 2015 that there are no essential sources of $O(^{1}D)$ in the mesopause during night (the minor sources: $O(^{1}S) \rightarrow O(^{1}D) + h\nu(557.7 \text{ nm}) \text{ and } O_{2}^{+} + e \rightarrow O(^{1}D) + O \text{ are}$ almost negligible). Recently, a new source of $O(^1D)$ population $\left(O+OH_{\nu\geq 5}\to O(^1D)+OH_{0\leq \nu^{'}\leq \nu-5}\right)$ was proposed (Sharma et al. 2015; Kalogerakis et al. 2016) as a hypothesis to explain the difference between experiment (Kalogerakis et al. 2011) and theory (e.g. Caridade et al. 2013, and references therein). It was successfully used to explain the deviations between numerical simulations and measurements of carbon dioxide 4.3 µm emission (Panka et al. 2017). Hence now, this mechanism is a wellestablished hypothesis. If this hypothesis is true, a new source for the production of $O_2(b^1\Sigma_g^+, \nu = 0)$ via $O(^1D)$ takes place in the mesopause region at night. In the present, there are different opinions about the canal $O + OH_{\nu=9} \rightarrow O(^{1}D) + OH_{\nu'=4}$. A number of authors takes this branch in account; on the other hand, there is an opinion that this canal does not exist (e.g. Yankovsky, private communication; Khomich et al. 2008). This still opened question does not influence all discussion below. Hence, in our manuscript, the original nomenclature (Sharma et al. 2015), which takes into an account this canal, is used.

Recently, by analysis of Energy Transfer in the Oxygen Nightglow (ETON 2) and Non-Local Thermodynamic Equilibrium (NLTE 2) rocket measurements, Kalogerakis (2019) inferred that the 762 nm emission via this new mechanism is over half of the total Atmospheric band emission near the peak, and may even exceed the emission from traditional source (two-step mechanism). Note, this conclusion, as well as discussion below, is valid and for the emission 865 nm which have as a source the same excited state of molecular oxygen.

Hence, a question arises regarding how is it possible that the fit-function, which was derived based on the assumption about the dominance of the two-step mechanism with empirically derived fitting coefficients (McDade et al. 1986; Grygalashvyly et al. 2019), has been used more than 30 years to infer atomic oxygen from observed Atmospheric band emission (e.g. Hedin et al. 2009), or vice versa, for simulations of emission from

atomic oxygen (e.g. Ward 1999) with well-confirmed results, if an additional strong mechanism exists.

In this short note, we show that in the case of Kalogerakis–Sharma Mechanism (KSM), the analogous fit-function can be derived. The analytical derivation is shown in the next section. The conclusions are summarised in the last section.

Analytical derivation

Assuming the two-step mechanism as dominant for $O_2\left(b^1\Sigma_g^+,\nu=0\right)$ population (hereafter, ν is the vibrational number for all vibrationally excited molecules), McDade et al. (1986) derive fit-function:

$$C^{O_2}[O_2] + C^{O}[O]$$

$$= \frac{A_{762}a_5[O]^2[O_2][M]}{V_{762}(A_{O_2(b_1)_0} + b_{O_2}[O_2] + b_{N_2}[N_2] + b_{O}[O])},$$
(1)

where a_5 is the reaction rate for atomic oxygen recombination; A_{762} is Einstein coefficient for Atmospheric band (762 nm) emission, V_{762} is corresponding volume emission; $A_{{\rm O}_2(b1)_0}$ is Einstein coefficient for total spontaneous emission of ${\rm O}_2\left(b^1\sum_g^+,\nu=0\right)$; $b_{{\rm O}_2},b_{{\rm N}_2},b_{{\rm O}}$ are the quenching rates of ${\rm O}_2\left(b^1\sum_g^+,\nu=0\right)$ with molecular oxygen, molecular nitrogen and atomic oxygen, respectively; and $C^{{\rm O}_2}$, $C^{{\rm O}}$ are the empirical fitting coefficients that were calculated from the ETON 2 rocket experiment. For more comfortable reading, we collect all nomenclature of reactions used in the manuscript in Table 1.

To show that in the case of KSM can be derived by analogous expression, we start from the expression for excited hydroxyl. Excited hydroxyl is in photochemical equilibrium in the mesopause region at night. Under this condition, its concentration is expressed as the ratio of productions to losses. The ozone is in the photochemical equilibrium in the vicinity of the excited hydroxyl layer and above at nighttime conditions (Belikovich et al. 2018; Kulikov et al. 2018, 2019). We use the expression for ozone balance at night $a_2[O][O_2][M] = a_1[O_3][H] + a_3[O][O_3]$, where a_1, a_2, a_3 are the coefficients for corresponding reactions (see Table 1). The reaction of ozone with atomic oxygen is relatively slow and can be omitted (Smith et al. 2008). Then, we substitute the reduced ozone balance equation $(a_2[O][O_2][M] = a_1[O_3][H])$ into the excited hydroxyl balance equation (first term in the numerator). Hence, we can write an equation where the concentration of excited hydroxyl is represented as a function of temperature (because of temperature-dependent reaction rates) and atomic oxygen concentration:

Table 1 List of reactions, nomenclature of reaction rates, branching ratios, quenching coefficients, spontaneous emission coefficients used in the paper

	Reactions	Coefficients/ branching ratios
R1	$H + O_3 \xrightarrow{\eta_V a_1} OH_{V=5,\dots,9} + O_2$	$a_1/\eta_{v=9,,5}$
R2	$O + O_2 + M \rightarrow O_3 + M$	a_2
R3	$O + O_3 \rightarrow 2O_2$	<i>a</i> ₃
R4	$O + OH_{v=5,,9} \rightarrow O_2 + H$	$a_4(v=9,\ldots,5)$
R5	$O + O + M \rightarrow O_2 + M$	a_5
R6	$O + OH_{v \ge 5} \xrightarrow{\psi_v a_{KSM}} O(^1D) + OH_{0 \le v' \le v - 5}$	$a_{\text{KSM}}/\psi_{v=9,\dots,5}$
R7	$O(^1D) \rightarrow O + hv$	A _{O(1D)}
R8	$O(^1D) + O_2, N_2, O \rightarrow \text{products}$	C_{O_2}, C_{N_2}, C_{O}
R9	$O(^1D) + O_2 \stackrel{\chi_0 c_{O_2}}{\rightarrow} O_2(b^1\Sigma_g^+, v = 0) + O$	X 0
R10	$O(^1D) + O_2 \xrightarrow{\chi_1 c_{O_2}} O_2(b^1\Sigma_q^+, v = 1) + O$	X 1
R11	$O_2(b^1\Sigma_g^+, v = 1) \rightarrow \text{products} + \text{hv(total)}$	$A_{O_2(b1)_{-1}}$
R12	$O_2(b^1\Sigma_g^+, v = 0) \rightarrow \text{products} + \text{hv(total)}$	$A_{O_2(b1)}_0$
R13	$O_2(b^1\Sigma_g^+, v = 0) \to O_2 + hv(762nm)$	A ₇₆₂
R14	$O_2(b^1\Sigma_g^+, v = 0) + O_2, N_2, O \rightarrow \text{products}$	$b_{\mathrm{O}_2}, b_{\mathrm{N}_2}, b_{\mathrm{O}}$
R15	$O_2(b^1\Sigma_g^+, v = 1) + O_2, N_2, O \to O_2$	$d_{\rm O_2}, d_{\rm N_2}, d_{\rm O}$
	$(b^1 \Sigma_g^+, v = 0) + O_2, N_2, O$	
R16	$OH_{v} + O_{2}, O, N_{2} \rightarrow OH_{v' < v} + O_{2}, O, N_{2}$	$Q_{vv^\prime}, P_{vv^\prime}, S_{vv^\prime}$
R17	$OH_{\nu} \rightarrow OH_{\nu' < \nu} + h\nu$	$E_{vv'}$

(R6). Taking into account that the most effective losses of excited hydroxyl occur by quenching with molecular oxygen, we can write simplified expressions for excited hydroxyl concentration (Grygalashvyly, 2015, eq. 14):

$$[OH_{\nu}] = \mu_{\nu} a_2 [O][M],$$
 (3)

where μ_{ν} are the coefficients which represent the arithmetic combination of branching ratios, η_{ν} and quenching coefficients $Q_{\nu'\nu}$ (collected in Appendix). Similar approach was used in a number of papers and is repeated here just for clarity (e.g. Mlynczak et al. 2014; Grygalashvyly et al. 2014; Grygalashvyly2015).

KSM populates $O(^1D)$ from vibrationally excited hydroxyl with $\nu = 5, ..., 9$ (Sharma et al. 2015; Kalogerakis et al. 2016; Panka et al. 2017; Kalogerakis 2019). $[O(^1D)]$ due to KSM using photochemical equilibrium (the life time is less than 1 s because of strong quenching; e.g. Slanger et al. (2017), Yankovsky and Manuilova (2018)) and can be written as follows:

$$\left[O(^{1}D)\right]_{KSM} = \frac{\sum_{\nu=5}^{\nu=9} \psi_{\nu} a_{KSM}[OH_{\nu}][O]}{A_{O(1D)} + c_{O_{2}}[O_{2}] + c_{N_{2}}[N_{2}] + c_{O}[O]},$$
(4)

where $A_{O(1D)}$ is the the total coefficient of $O(^1D)$ spontaneous emission, and c_{O_2} , c_{N_2} , c_O are the quenching coefficients for deactivation of $O(^1D)$ by molecular oxygen, molecular nitrogen, and atomic oxygen, respectively.

Two lowest vibrational states of $O_2(b^1\Sigma_g^+, \nu = 0.1)$ are populated via quenching of $O(^1D)$ by molecular oxygen, with branching ratios χ_0 and χ_1 , respectively. Then,

$$[OH_{\nu=9,...,5}] = \frac{\begin{pmatrix} \eta_{\nu}a_{2}[O][O_{2}][M] + \sum_{\nu'=\nu+1}^{9} P_{\nu'\nu}[OH_{\nu'}][O] + \sum_{\nu'=\nu+1}^{9} S_{\nu'\nu}[OH_{\nu'}][N_{2}] + \\ + \sum_{\nu'=\nu+1}^{9} Q_{\nu'\nu}[OH_{\nu'}][O_{2}] + \sum_{\nu'=\nu+1}^{9} E_{\nu'\nu}[OH_{\nu'}] \end{pmatrix}}{\begin{pmatrix} a_{4}(\nu)[O] + \sum_{\nu''=0}^{\nu-1} P_{\nu\nu''}[O] + \sum_{\nu''=0}^{\nu-1} S_{\nu\nu''}[N_{2}] + \\ + \sum_{\nu''=0}^{\nu-1} Q_{\nu\nu''}[O_{2}] + \sum_{\nu''=0}^{\nu-1} E_{\nu\nu''} + \psi_{\nu}a_{KSM}[O] \end{pmatrix}}, \begin{pmatrix} \nu < \nu' \\ \nu'' < \nu \end{pmatrix}.$$
(2)

Here, η_{ν} are the branching ratios for the reaction of atomic hydrogen with ozone; a_2 , a_4 are the reaction rates for R2 and R4, respectively; $Q_{\nu'\nu}$, $P_{\nu'\nu}$, $S_{\nu'\nu}$ are the quenching coefficients (where lower indexes denote corresponding transitions) for deactivation of excited hydroxyl by molecular oxygen, atomic oxygen, and molecular nitrogen, respectively; $E_{\nu'\nu}$ are the spontaneous emission coefficients; a_{KSM} is the reaction rate for KSM, and ψ_{ν} are the branching ratios for KSM. Note, in the denominator of Eq. (2), we take into an account not only quenching processes and spontaneous emission, but, additionally, chemical removing (following by terminology of e.g. Varandas (2004), Caridade et al. (2013), reactive quenching), and removing by KSM. Hence, we separate three different processes: non-reactive quenching (R16), reactive quenching (R4), and Kalogerakis-Sharma process $O_2(b^1\Sigma_g^+, \nu=1)$ can be deactivated by quenching with O_2 , N_2 , and O into zero vibrational state, or it can be transformed into ground state via spontaneous emission (e.g. Pejakovic 2014; Yankovsky et al. 2016, 2019).

 ${\rm O}_2\Big(b^1\Sigma_g^+, \nu=0\Big)$ is the result (at nighttime conditions) of deactivation of O(^1D) by molecular oxygen and ${\rm O}_2\Big(b^1\Sigma_g^+, \nu=1\Big)$ by O₂, N₂, and O. Then, it is deactivated in processes of spontaneous emission and quenching.

The radiative lifetimes of $O_2(b^1\Sigma_g^+, \nu=0.1)$ are less than 12 s (e.g. Yankovsky et al. 2016, 2019 and references therein), hence, assuming photochemical equilibrium for both, we can express them as a ratio of production to the losses.

$$\left[O_{2}\left(b^{1}\Sigma_{g}^{+}, \nu = 1\right)\right]_{\text{KSM}} = \frac{\chi_{1}c_{\text{CO}_{2}}[O_{2}]\left[O^{1}D\right]_{\text{KSM}}}{\left(A_{O_{2}(b1)} + d_{O_{2}}[O_{2}] + d_{N_{2}}[N_{2}] + d_{O}[O]\right)},\tag{5}$$

where $A_{\mathrm{O}_2(b1)_-1}$ is the Einstein coefficient for total spontaneous emission of $\mathrm{O}_2\Big(b^1\Sigma_g^+,\nu=1\Big)$; χ_1 is branching ratio (see above); and $d_{\mathrm{O}_2},d_{\mathrm{N}_2},d_{\mathrm{O}}$ are the rates of quenching of $\mathrm{O}_2\Big(b^1\Sigma_g^+,\nu=1\Big)$ by molecular oxygen, molecular nitrogen, and atomic oxygen, respectively.

where
$$C^{O_2} = \frac{a_5}{\chi a_2 a_{KSM} \sum_{\nu=5}^{\nu=9} \psi_{\nu} \mu_{\nu}} \left(1 + \frac{c_{N_2}[N_2]}{c_{O_2}[O_2]} \right)$$
 and $C^O = \frac{c_{Oa_5}}{\chi a_2 a_{KSM} c_{O_2} \sum_{\nu=9}^{\nu=9} \psi_{\nu} \mu_{\nu}}$.

Hence, the case of leading KSM gives the same expression for fit-function.

$$\label{eq:continuous_continuous$$

where χ_0 is branching ratio (see above); $b_{\rm O_2}, b_{\rm N_2}, b_{\rm O}$ are the quenching rates of ${\rm O_2}\left(b^1\Sigma_g^+, \nu=0\right)$ with molecular oxygen, molecular nitrogen and atomic oxygen, respectively.

Next, we substitute (5) into (6). The spontaneous emission coefficient $A_{O_2(b_1)_{-1}}$ is much smaller than other terms in the denominator of (5) (Pejakovic et al. 2005a,b). Hence, we can write

Conclusions

The fit-functions for KSM and for two-step mechanism have analogous analytical expressions. This explains why a long time of using this expression with empirically derived fitting coefficients but without detailed knowledge on acting mechanisms gives approximately correct values of atomic oxygen and Atmospheric band volume emissions. In the frame of our analytical approach

$$\left[O_{2}\left(b^{1}\Sigma_{g}^{+}, \nu = 0\right)\right]_{\text{KSM}} = \frac{\chi c_{O_{2}}[O_{2}]\left[O^{(1}D)\right]_{\text{KSM}}}{\left(A_{O_{2}(b_{1})} = 0 + b_{O_{2}}[O_{2}] + b_{N_{2}}[N_{2}] + b_{O}[O]\right)},\tag{7}$$

where $\chi=\chi_0+\chi_1$ is the total branching ratio for ${\rm O}_2\Big(b^1\Sigma_g^+,\nu=0,1\Big)$ population.

The KSM which produces $O(^1D)$, and consecutively $O_2(b^1\Sigma_g^+, \nu=0)$, is responsible for part of Atmospheric band emission (762 nm):

$$V_{\text{KSM}} = A_{762} \Big[O_2 \Big(b^1 \Sigma_g^+, \nu = 0 \Big) \Big]_{\text{KSM}}.$$
 (8)

Substituting (3) into (4), then (4) into (7), and finally (7) into (8), we obtain:

and currently available rocket experiments, we do not have the possibility to make any solid conclusions about the balance between two mechanisms. To highlight this problem, new simultaneous observations of the Atmospheric band emission, excited hydroxyl emissions, atomic oxygen and temperature are necessary. Additionally, laboratory investigations to study the branching ratios and reaction rate for KSM are desirable.

$$V_{\text{KSM}} = \frac{A_{762} \chi c_{\text{O}_2} a_{\text{KSM}} a_2[\text{O}]^2[\text{O}_2][\text{M}] \sum_{\nu=5}^{\nu=9} \psi_{\nu} \mu_{\nu}}{\left(A_{\text{O}_2(b1)_0} + b_{\text{O}_2}[\text{O}_2] + b_{\text{N}_2}[\text{N}_2] + b_{\text{O}}[\text{O}]\right) \left(A_{\text{O}(1\text{D})} + c_{\text{O}_2}[\text{O}_2] + c_{\text{N}_2}[\text{N}_2] + c_{\text{O}}[\text{O}]\right)}.$$
(9)

Omitting emissive term $A_{O(1D)}$ as non-effective loss and reorganising (9), we obtain

$$C^{O_2}[O_2] + C^{O}[O] = \frac{A_{762}a_5[O]^2[M][O_2]}{V_{KSM}(A_{O_2(b_1)} + b_{O_2}[O_2] + b_{N_2}[N_2] + b_{O}[O])},$$
(10)

Abbreviations

KSM: Kalogerakis-Sharma Mechanism; PWs: Planetary waves; GWs: Gravity waves; ETON: Energy Transfer in the Oxygen Nightglow; NLTE: Non-Local Thermodynamic Equilibrium.

$\mu_8 = \frac{\eta_8}{O_8} + \frac{\eta_9 Q_{98}}{O_9 O_8}$

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$$\mu_7 = \frac{\eta_7}{Q_7} + \frac{\eta_9 Q_{97}}{Q_9 Q_7} + \frac{\eta_8 Q_{87}}{Q_8 Q_7} + \frac{\eta_9 Q_{98} Q_{87}}{Q_9 Q_8 Q_7},$$

$$\mu_{6} = \frac{\eta_{6}}{Q_{6}} + \frac{\eta_{9}Q_{96}}{Q_{9}Q_{6}} + \frac{\eta_{8}Q_{86}}{Q_{8}Q_{6}} + \frac{\eta_{9}Q_{98}Q_{86}}{Q_{9}Q_{8}Q_{6}} + \frac{\eta_{7}Q_{76}}{Q_{7}Q_{6}} + \frac{\eta_{9}Q_{97}Q_{76}}{Q_{9}Q_{7}Q_{6}} + \frac{\eta_{8}Q_{87}Q_{76}}{Q_{8}Q_{7}Q_{6}} + \frac{\eta_{9}Q_{98}Q_{87}Q_{76}}{Q_{9}Q_{7}Q_{6}},$$

$$\begin{split} \mu_5 &= \frac{\eta_5}{Q_5} + \frac{\eta_9 Q_{95}}{Q_9 Q_5} + \frac{\eta_8 Q_{85}}{Q_8 Q_5} + \frac{\eta_9 Q_{98} Q_{85}}{Q_9 Q_8 Q_5} + \frac{\eta_7 Q_{75}}{Q_7 Q_5} + \frac{\eta_9 Q_{97} Q_{75}}{Q_9 Q_7 Q_5} + \frac{\eta_8 Q_{87} Q_{75}}{Q_8 Q_7 Q_5} + \frac{\eta_9 Q_{98} Q_{87} Q_{75}}{Q_9 Q_8 Q_7 Q_5} + \\ &+ \frac{\eta_6 Q_{65}}{Q_6 Q_5} + \frac{\eta_9 Q_{96} Q_{65}}{Q_9 Q_6 Q_5} + \frac{\eta_8 Q_{86} Q_{65}}{Q_8 Q_6 Q_5} + \frac{\eta_9 Q_{98} Q_{86} Q_{65}}{Q_9 Q_8 Q_6 Q_5} + \frac{\eta_7 Q_{76} Q_{65}}{Q_7 Q_6 Q_5} + \frac{\eta_9 Q_{97} Q_{76} Q_{65}}{Q_9 Q_7 Q_6 Q_5} + \\ &+ \frac{\eta_8 Q_{87} Q_{76} Q_{65}}{Q_8 Q_7 Q_6 Q_5} + \frac{\eta_9 Q_{98} Q_{87} Q_{76} Q_{65}}{Q_9 Q_8 Q_7 Q_6 Q_5}. \end{split}$$

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Competing interests

The authors declare that they have no conflict of interest.

Appendix

Coefficients μ_{ν} for Eq. (3). Here $Q_{\nu} = \sum_{\nu'=\nu-1}^{\nu'=\nu-1} Q_{\nu\nu'}$ is the sum of quenching ratios for OH_{ν} by O_2 for transitions into all lower vibrational levels.

$$\mu_9 = \frac{n_9}{Q_9},$$

On the other hand, these coefficients can be represented by the simple recursive expression:

$$\mu_{v} = \frac{\eta_{v} + \sum_{v'=v+1}^{v'=9} \mu_{v'} Q_{v'v}}{\sum_{v''=0}^{v''=v-1} Q_{vv''}}, (\eta_{v>9} = 0).$$

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