UPPER ATMOSPHERE DAYGLOW EMISSIONS at $\lambda = 589$, 330, and 285 nm IN THE PRESENCE OF NaCl VAPOR

A.M. Samusenko

Institute of Physics, Academy of Sciences of the Bellorussian SSR, Minsk Received April 9, 1990

The mechanism of Na dayglow emission in the upper atmosphere ($z \ge 80$ km) is studied. The Na is formed as a result of photodissociation of man-made NaCl vapor. It is shown that for relative NaCl concentration of the order of 10^{-4} this mechanism gives intense emission in the resonance lines of Na in the disturbed region of space.

Two chemiluminescence mechanisms of mesosphere nightglow, produced when jet-engine exhaust interacts with the surrounding atmosphere, are studied in Refs. 1 and 2. Because of these processes at night at altitudes of z = 80-100 km the intensity of hydroxyl emission and sodium D-line emission in the wake (the region where the products of combustion are mixed with the components of the atmosphere) can be one to two orders of magnitude higher than in the undisturbed atmosphere.

In studying the daytime emission of the wake these mechanisms are not important during the daytime the chemiluminescence emission is much stronger than the background light. However when the wake, containing the same components as in Ref. 2, is irradiated by the sunlight excited Na can form by a different (and very efficient) mechanism. The intensity of the Na emission is much stronger than the background light not only at the wavelength 589 nm, but also at $\lambda = 330$ and 285 nm. The formulation and method of investigation of this problem are similar to Refs. 1 and 2. For this reason we shall discuss below primarily the differences between the dayglow and nightglow of Na in the wake.

One of the main differences is that during the daytime the region of sodium emission has only a lower boundary z = 80 km. As in Ref. 2 the existence of a lower boundary is connected with the existence of sodium sink in which sodium is transformed into chemically inert NaO₂ via the reaction 2 in Table I.

With the exception of the reaction 9, Table I is identical to the analogous table for nightglow.² But the process 9 radically changes the situation, since now the main source of atomic Na in the wake is photodissociation of NaCl. This is the second important difference between dayglow and nightglow. In addition, at night the formation of atomic sodium is, in principle, impossible without the reactions 1 and 3 (this, by the way, is why the altitude of the region of chemiluminescence has an upper bound), whereas during the daytime the reaction 9 by itself is sufficient for Na to appear in the wake at any altitude above 80 km. The fraction of atomic Na that is converted directly via the reactions 1-8 into other sodium compounds decreases as the altitude increases and starting at z = 100 km, because the processes 1 and 2 play a negligible role here, virtually none of the Na formed by photodissociation of NaCl is chemically transformed.

TABLE 1. Basic reactions involving sodium in the daytime wake at altitudes of 80–100 km.

Reaction	K, cm ⁻³ s ⁻¹	τ, s
1. Na + $0_3 \rightarrow NaO + 0_2$	3.1·10 ⁻¹⁰	20
2. Na + O_2 + $N_2 \stackrel{\beta}{\rightarrow} O_2$ + N_2	6.7·10 ⁻³¹ e ^{290/T}	10-10 ³
3a. NaO + O \rightarrow Na + O 2	3 7.10 -10	0.005-0.2
3b. NaO + O \rightarrow Na(² P) + O ₂ \int	5.7 10	0.000 0.2
4. NaO + $H_2O \rightarrow NaOH + OH$	2.2·10 ⁻¹⁰	0.001-0.01
5a. NaO + $H_2 \rightarrow NaOH + H$	2.6.10-11	0.005-0.05
5b. NaO + $H_2 \rightarrow Na + H_2O$	$\frac{k_{5b} + k_{5c}}{k_{5}} > 0.2$	
5c. NaO + $H_2 \rightarrow Na(^2P) + H_2O$	-	
6. NaO + HCl \rightarrow NaCl + OH	2.10-10	0.001-0.01
7. NaOH + HCl \rightarrow NaCl + H ₂ O	2.10-10	0.001-0.01
8. NaO_2 + HCl \rightarrow NaCl + HO ₂	2.3.10-10	0.001-0.01
9. NaCl + $h\nu \rightarrow Na$ + Cl	(1-2).10 ⁻³	~10 ³

Note.

1. The quantities β and J^{∞} have the dimensions cm⁻⁶ · s⁻¹ and s⁻¹, respectively. The rate J^{∞} was estimated from data on the photodissociation cross sections³ for radiation flux incident perpendicular on the boundary of the atmosphere when the Sun is quiet (see, for example, Ref. 4).

2. The quantities τ_{chem} are the characteristic times over which the concentrations of reagents containing Na change.

3. The typical composition of jet exhaust includes H_2 , H_2O , CO, CO_2 , HCl, as well as traces of metals and their compounds (Na, NaCl, etc.).²

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The third significant difference is that the reaction times change. At night these times range from fractions of a second to one second. The photodissociation time of NaCl ($J_9^{-1} \sim 10^3$ s) determines the dayglow. Thus in Refs. 1 and 2 it was sufficient to consider the initial stage of wake expansion (i.e., a simple relation of the form $R^2 \sim t$ between the radius of the wake R and the time t was sufficient). In our problem, however, wake evolution must be studied over a longer period of time. In particular, the facts that in time wake expansion slows down and the wake stabilizes and changes only as a result of exoatmospheric processes and molecular diffusion must be taken into account.

Finally, the last difference is connected with the mechanism of excitation of Na^{*}. At night this is simply the chemiluminescence mechanism, but during the day resonance fluorescence plays the main role. It is because of excitation by solar radiation that Na can emit in the wake not only at 589 nm but also at the shorter wavelengths 330 and 285 nm.

We shall formulate the kinetic equations not for the concentrations of the reagents, but rather for their integrated quantity

$$dz \int_{0}^{R(t)} [X] 2\pi r dr = dz \{X\}$$
(1)

([X] is the concentration of the chemical element X and r is the running value of the radial coordinate in the wake) in a layer of thickness dz of a vertically oriented cylindrically symmetric wake.^{1,2} This simplified approach makes it possible to ignore the effect of dynamic processes on the change in [X]. The price for this simplification is that ultimately only the average values of the concentrations can be determined according to the formula

$$\overline{[X]} = \{X\}/\pi R^2(t) .$$
(2)

Based on the kinetic scheme presented in Table I, with the help of Eqs. (1) and (2) we obtain

$$\frac{d\{Na0\}}{dt} = k_1[0_3] \{Na\} - [k_3[0] + k_4[H_20] + k_5[H_2] + k_6[HC1]]\{Na0\};$$

$$\frac{d\{Na\}}{dt} = -k_1[0_3] \{Na\} - k_2[0_2] [M] \{Na\} + [Na0\} (k_3[0] + 0.2 k_5[H_2]) + J_9^{(0)}\{NaC1\};$$

$$\frac{d\{Na0\}}{dt} = k_2[0_2] [M] \{Na\} - k_6[HC1] \{Na0_2\};$$

$$\frac{d\{Na0H\}}{dt} = k_4 [H_20] \{Na0\} + 0.8k_5 [H_2] \{Na0\} - k_7 [HC1] \{Na0H\};$$

$$\frac{d\{NaC1\}}{dt} = -J_9^{(0)}\{NaC1\} + (k_6\{Na0\} + k_7\{Na0H\} + k_8\{Na0_2\}) [HC1].$$
(3)

The system (3) is complicated, even if the dynamical terms are neglected. Under certain conditions, however, this system can be simplified. It evident from Table I that the chemical is transformation times of the compounds NaO, NaO₂, and NaOH are equal to fractions of a second. These times are extremely short compared with the characteristic time of the problem $J^{-1} \sim 10^3$ s. It is obvious that for times t comparable to J^{-1} the quantities {NaO}, {NaO2}, and {NaOH} will be quasistationary, i.e., in the corresponding differential equations the derivatives with respect to t can be set equal to zero. Ultimately three of the five equations of the system (3) transform into algebraic relations and the system (3) reduces to a system of two equations:

$$\frac{d\{\operatorname{Na}\}}{dt} = -f(t) \{\operatorname{Na}\} + J_{g}\{\operatorname{NaC1}\};$$

$$\frac{d\{\operatorname{NaC1}\}}{dt} = -J_{g}\{\operatorname{NaC1}\} + f(t) \{\operatorname{Na}\}, \qquad (4)$$

where

$$f(t) = \frac{\alpha k_1 [0_3]}{k_3 [0_3] \frac{R^2(t)}{R_0^2} + \beta_0} + k_2 [0_2] [M] ,$$

$$\alpha = k_4 \overline{[H_20]}_0 + 0.8 k_5 \overline{[H_2]}_0 + k_6 \overline{[HC1]}_0 ,$$

$$\beta = \alpha + 0.2 k_5 \overline{[H_2]}_0, \qquad J_9 = J_9^{\infty} .$$
(5)

The index "0" designates the initial values of the wake parameters.

For what follows we must specify how the wake expands with time. The dependence $R^2(t)$ employed In Refs. 1 and 2 is an approximation of the numerical calculations of Ref. 7. It is correct only at the initial stage of wake expansion, and it is based on the assumption that turbulent mixing, assumed in Ref. 7, occurs up to z = 100 km. Extrapolation to altitudes above 100 km is not justified because at those altitudes other mechanisms of mixing predominate.

The assumption that at altitudes $z \leq 100$ km turbulent mixing determines wake formation is used in this paper also. However, it should be kept in mind that during the daytime it is not enough to study only the initial stage of wake expansion. The characteristic photodissociation time of NaCl is ~ 10^3 s, and therefore the concentration of the source of fluorescence — atomic sodium — reaches its highest values at times $t \gg 100$ s.

In Ref. 8, where the ideas incorporated in Ref. 7 are developed, it is shown by direct numerical calculations that the one-dimensional approximation satisfactorily describes the mixing not only at the early stages but also at the later stages of evolution of the wake (in the calculations the time ran from 0 to 600 s). The analytical formulas themselves are not convenient for calculations, but analysis of the formulas shows that at least for $t \leq 600$ s two characteristic wake-expansion times can be separated: for $t \leq 250$ s the radius of the wake increases as $R^2(t) \sim t$ with characteristic time of the order of several seconds, while for t > 250 s $R^2(t)$ grows with a characteristic time of hundreds of seconds.

The approximation

$$R^{2}(t)/R_{0}^{2} = \left[1 + \frac{\tau_{\bullet}}{\tau_{D}} \left(1 - \exp(-t/\tau_{\bullet})\right)\right],$$

$$R_{0}^{2} = ae^{bz}$$
(6)

with the parameters $\tau_d = 2.5$ s, $\tau_* = 500$ s, a = 1.4 m, and $b = 5 \cdot 10^{-5}$ m satisfactorily describes the results of Refs. 7 and 8; it agrees well with Refs. 1 and 2 for t < 100 s; and, it also describes qualitatively correctly wake evolution at long times ($t > 10^3$ s; atmospheric processes are neglected). Indeed, if dynamic processes in the upper atmosphere are neglected, then for $t > 10^3$ s the wake should be stable and its parameters should vary slowly because molecular diffusion.

Using Eqs. (5) and (6) we shall find an approximate solution of the system (4)

$$\times \ln \left[\frac{(1+\gamma) \exp(t/\tau_{*}) - 1}{\gamma} \right] + \frac{J_{9}}{a_{2} + J_{9} + \frac{\alpha a_{1}\tau_{p}}{a_{3}\tau_{*}(1+\gamma)}} \left[\{ Na \}_{0} + \{ NaCl \}_{0} \} \times \left[1 - \exp \left[- \left[a_{2} + J_{9} + \frac{\alpha a_{1}\tau_{p}}{a_{3}\tau_{*}(1+\gamma)} \right] t \right] \right]$$
(7)

where

$$\begin{aligned} a_1 &\equiv k_1 \begin{bmatrix} 0_3 \end{bmatrix}, \quad a_2 &\equiv k_2 \begin{bmatrix} 0_2 \end{bmatrix} \begin{bmatrix} M \end{bmatrix}, \quad a_3 &\equiv k_3 \begin{bmatrix} 0 \end{bmatrix}, \\ \gamma &\equiv \frac{\beta \tau_{\rm D}}{a_3 \tau_{\bullet}}, \quad \beta &\equiv \beta_0 + a_3. \end{aligned}$$

The first term in Eq. (7) describes the loss of Na present initially in the exhaust. The second term describes the production of Na by photodissociation of NaCl. Since a_2 depends quadratically on the pressure, at the bottom boundary (z = 80 km) the first term in Eq. (7) becomes vanishingly small already at times $t \sim 10$ s. Over the same time the second term reaches a stationary value of $\simeq \frac{J_9}{a_2}$ {NaCl}₀ (in the exhaust {NaCl}₀ \gg {Na}₀).^S

TABLE II. The values of the concentrations [Na] in cm^{-3} .

a km	t, s								
2, Km	0**	50	100	250	500	750	1000	2000	5000
80	1.0+8	3.4+6	2.0+6	9.1+5	5.7+5	4.7+5	4.2+5	3.7+5	3.7+5
82.5	8.4+7	6.8+6	4.0+6	1.8+6	1.1+6	9.4+5	8.4+5	7.4+5	7.4+5
85	7.2+7	1.1+7	7.2+6	3.6+6	2.3+6	1.8+6	1.7+6	1.5+6	1.5+6
87.5	5.8+7	1.1+7	9.1+6	5.7+6	3.8+6	3.1+6	2.8+6	2.4+6	2.4+6
90	4.2+7	1.0+7	9.1+6	7.2+6	5.7+6	4.9+6	4.4+6	3.9+6	3.9+6
92.5	3.5+7	8.9+6	8.3+6	7.4+6	6.6+6	6.1+6	6.0+6	5.3+6	5.3+6
95	2.8+7	7.7+6	7.4+6	7.0+6	7.3+6	7.4+6	7.5+6	7.7+6	7.8+6
97.5	2.2+7	6.3+6	5.9+6	6.1+6	6.4+6	6.9+6	7.3+6	8.4+6	9.1+6
100	1.5+7	4.3+6	4.1+6	4.1+6	4.5+6	4.9+6	5.2+6	6.1+6	6.8+6

* $1.0+8 = 1.0 \cdot 10^{+8}$.

** The initial distribution of [Na]₀.

Because at 80 km $J_9/a_2 \ll 1$, however, only a smallfraction of NaCl present is converted into sodium. The ratio J_9/a_2 is close to unity only at altitudes $z \ge 95$ km. For this reason, .starting at altitudes ~ 95 km virtually all of the NaCl is converted in Na, but unlike at z = 80 km the conversion time J_9^{-1} is now ~ 500–1000 s.

We shall calculate with the help of Eqs. (2), (6), and (7) the average sodium concentrations in the wake for $z \in 80\text{-}1000$ km. We shall assume that the relative concentration of Na and NaCl in the exhaust is the same as in Ref. 5: $C_{\text{Na}} = 2 \cdot 10^{-6}$, $C_{\text{NaCl}} = 2 \cdot 10^{-4}$. The initial altitude distribution of [Na]₀ with $C_{\text{Na}} = 2 \cdot 10^{-6}$ is given in Ref. 2. Using the ratio $C_{\rm NaCl} / C_{\rm Na} = 100$, it is also easy to obtain from the distribution of [Na]₀ the distribution of [NaCl]₀. The average concentrations [Na] calculated for the parameters given above are presented in Table II for a wide range of times.

It should be noted that it is possible to have a situation in which at $z \in 80\text{-}1000$ km all of the ozone mixed into the stream reacts within a time ~ 100 s after the airplane passes by.¹ For the present problem this fact, however, is not significant. According to Table I and the structure of the first term in Eq. (7), the lack of ozone will only decrease the rate of loss of {Na}₀, and because {NaCl}₀ \gg {Na}₀ this will have virtually no effect on the value of {Na}.

For z > 100 km the formula (6) is not applicable. At such altitudes, of the mechanisms presented in Table I only the photodissoclation of NaCl must be taken into account. Correspondingly, to find the total number of Na atoms in a region of the wake of thickness dz we must set f(t) = 0 in the system Eq. (4). Proceeding in this manner, for z > 100 km we find

{Na} = {Na}₀ + {NaCl}₀
$$\left[1 - e^{-Je}\right]$$
. (8)

Unfortunately, the value of $\{Na\}$ permits determining only the integrated energy characteristics of the emission, but not a differential property, for example, the intensity I.

The solar radiation incident on a wake containing Na is absorbed in one of the resonance lines and then reradiated in all directions within the same line. At the altitudes of interest the collisional quenching of electronically excited states of Na can be neglected. For this reason, the power of the scattered radiation intensity will be equal to the absorbed radiation intensity, and in this sense the latter can be used to describe the emission of the disturbed atmosphere. Nonetheless a simple relation between the emission and absorption exists only for an optically thin, medium.

We shall find with the help of the data in Table II the optical thicknesses of the wake at $\lambda = 589, 330$, and 285 nm (the transitions $3S \rightarrow 3P$, $3S \rightarrow 4P$, and $3S \rightarrow 5P$, respectively) for different altitudes z and times t in the case when the vertically oriented wake is illuminated by perpendicular rays of the sun. The optical thickness along the path l is calculated with the help of the formula

$$\tau = \sigma_0 [\text{Na}] l , \qquad (9)$$

where σ_0 is the absorption cross section at the center of the line. Substituting into Eq. (9) the corresponding values of the parameters of the problem, it can be shown that for l = 2R and $z > 82.5 \text{ km } \tau_{589}$ is much greater than unity for all values of [Na] presented in Table II. At the two other wavelengths the optical thicknesses are much less than unity for all t and z. This makes it possible to use the volume approximation when calculating I_{330} and I_{285} . Because of the strong inequality $\tau \gg 1$ the intensity of the outgoing radiation in the sodium *D*-line must be calculated using the transfer equation. This is a separate problem and falls outside the scope of this paper. Our purpose is not so much to obtain the detailed characteristics of the scattered light as it is to call attention to the importance of the effect of scattering of solar radiation by Na present in the wake. To this end it is sufficient to calculate the intensity of the absorbed energy I_{589} .

In the general case

$$I_{\nu}^{abs} = \omega_{eqv} F_{\nu}, \qquad (10)$$

where ω_{eqv} is the equivalent linewidth and F_v is the solar flux. For Doppler broadening⁹

$$w_{eqv} = 2\gamma_{D} \left[\frac{\ln\left(1 + 1.772f^{\bullet}L_{\bullet}\right)}{0.693f^{\bullet}\left(1 + \frac{2.26}{L_{\bullet}}\right)} \right]^{1/2},$$

$$f^{\bullet} \equiv \frac{0.2 + 0.3L_{\bullet}}{1 + 0.3L_{\bullet}}, \quad L_{\bullet} = 0.47\sigma_{0}\overline{[Na]} l,$$
(11)

where γ_D is the Doppler half-width of the line. Substituting into Eqs. (10) and (11) the parameters of the sodium *D*-line ($\gamma_D = 13 \cdot 10^9 \text{ s}^{-1}$, $\sigma_0^{589,0} = 2$, $\sigma_0^{589,6} =$ $= 1,3 \cdot 10^{-11} \text{ cm}^2$) and setting $F_{589}^{\infty} = 2 \cdot 10^{-16} \times$ $\times \text{ W/cm}^2 \cdot \text{ Hz}$ (the solar flux when the Sun is quiet),⁴ the data in Table II can be used to determine the intensity of the solar radiation absorbed in 1 cm² of the wake in each line of the doublet. For the 589.0 nm line the values of I^{abs} are presented in Table III. The intensities in the lines of the doublet are related with one another by the relation $I_{589,0} = 2I_{589.6}$.

It is evident that from Table III that over a long period of time $I^{abs} \simeq 10^{-6}$ W/cm² throughout virtually the entire altitude range 80–100 km. Since all of the absorbed energy is reradiated, the intensity of the scattered light summed over all directions is also approximately equal to 10^{-6} W/cm² $\simeq 3MR$. Thus the presence of the order of 0.01% NaCl in the exhaust results in strong wake emission at 589 nm.

For narrow lines, which, as we have pointed out, the lines at $\lambda = 330$ and 285 nm are, the intensity of the scattered radiation can be found immediately:

$$I_{\nu}^{\text{scat}} = \frac{h\nu}{4\pi} \int_{1}^{\varepsilon} \varepsilon \, \mathrm{d}l \quad , \tag{12}$$

$$\varepsilon = \sigma_0 \gamma_D F_v \overline{[Na]} . \tag{13}$$

Using the values of the parameters of the corresponding lines and the data in Table II, for z = 85-100 km and $t \le 5000$ s, we find that in the brightest part of the wake, i.e., for l = 2R(t), $I_{330}^{sc} \simeq (20-100)kR$ and

 $I_{285}^{sc} \simeq 400-2000R$. The intensity along any ray tangent to a circle of radius *r* centered on the axis of

the cylinder is $(1 - r^2/R^2)^{1/2}$ times smaller than the values presented.

TABLE III. The solar radiation power absorbed in 1 cm² of the wake at 589.0 nm (in units of 10^{-7} W/cm²).

- Im	t, s								
2, km	0	50	100	250	500	750	1000	2000	5000
80	10.7	5.3	4.4	3.4	2.8	2.6	2.5	2.3	2.3
82.5	10.6	8.0	7.1	5.8	5.0	4.8	4.7	4.4	4.4
85	10.5	9.7	9.2	8.4	7.8	7.4	7.4	7.2	7.2
87.5	10.3	9.9	10.1	9.9	9.6	9.2	9.2	9.0	9.0
90	9.9	9.9	10.7	10.9	10.9	10.9	10.7	10.7	10.7
92.5	9.7	9.9	10.7	11.1	11.3	11.6	11.6	11.3	11.3
95	9.5	9.9	10.7	11.1	12.0	12.2	12.2	12.4	12.4
97.5	9.2	9.8	10.3	11.1	12.0	12.2	12.4	12.8	13.0
100	8.4	9.2	9.8	10.7	11.3	12.0	12.2	12.4	12.6

Wake emission at altitudes z > 100 km cannot be investigated in detail because here the time dependence of the wake expansion is not known. The formula (8) in the case of an optically thin medium permits determining only the integrated characteristics of the emission, such as, for example, the total spectral energy E_v of the radiation from a section of the wake of unit length:

$$E_{\nu} = h\nu \int_{0}^{R} \varepsilon 2\pi r \, dr =$$

= $h\nu \sigma_{0} \gamma_{p} F_{\nu} [\{Na\}_{0}^{+} \{NaCl\}_{0} \left[1 - e^{-Jg^{t}}\right].$ (14)

Equation (14) was derived with the help of the expressions (8) and (13). If the radius of the wake is known, then the maximum (i.e., along the radius R) intensity in all directions can be estimated quite

simply using the formula $I_v = \frac{2}{\pi} E_v / R$.

Owing to the ratio $C_{\text{NaCl}}/C_{\text{Na}} = 100$, the energy of the radiation from a wake of unit length over a time $t \sim J_9^{-1} \sim 10^3$ s increases by a factor of 100 for $\tau < 1$. According to our model the quantity {NaCl}₀ for 1 cm of the wake is equal to $\sim 2 \cdot 10^{17}$ molecules. Based on this fact, the maximum values of E_v are as follows:

λ(nm)	589	330	285
$E_{v}\left(\frac{W}{cm}\right)$	8.6	2.5·10 ⁻²	8.3·10 ⁻⁴

For an optically thick wake (in the *D*-lines of Na such a situation also occurs for z > 100 km) the formula (14) is not applicable. In this case, however, the quantity *I* will be virtually identical to the values of the intensity presented in Table III. The reason lies

in the fact that for Doppler broadening the quantity ω_{eqv} for $\tau > 1$ is virtually independent of the optical thickness.

Thus we have shown that at high altitudes the ejection of Na or NaCl into the atmosphere can cause the optical characteristics of the atmosphere to change significantly. A disturbance of the atmospheric emission during the day lasts for a much longer period of time than at night, when the emission of Na or NaCl affects the atmospheric emission for a comparatively short period of time (~ 100 s after the aircraft passes by²). The question of the airglow duration can be solved only by considering additional mechanisms connected with atmospheric processes. Such processes, by mixing the disturbed and undisturbed parts of the atmosphere, reduce the nonuniformity of the chemical composition and, as a consequence, decrease I_v .

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