INFLUENCE OF AEROSOL ON THE TEMPERATURE DEPENDENCE OF WATER VAPOR CONTINUUM IN THE 8–12 μm RANGE

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Submicron aerosol, whose extinction coefficient in the visible spectral range depends on the air relative humidity, is shown to cause an essential overestimation of the negative temperature dependence of the water vapor absorption continuum in the 8–12 μm *range. This conclusion was drawn based on the experimental data [V.N. Aref'ev and N.I. Sizov, Tr. Inst. Eksp. Meteorol., Akad. Nauk SSSR, Obninsk, No. 14(110), 93–99 (1984)].*

When investigating light absorption by water vapor within the absorption continuum in the 8–12 μm transmission window under both laboratory and natural conditions there is a need for a correct exclusion of the aerosol extinction component. The neglect of this component can result, on the one hand, in an evident overestimation of the continuum absorption and, on the other hand, in the distortion of its temperature dependence.

To explain the second statement the following reasoning can be suggested.

It is known that the aerosol extinction increases with the increase of relative humidity of the air. As follows from the investigation of the dependence of aerosol extinction coefficient on the air absolute humidity at some fixed values of temperature (as is done in studies of the coefficients of continuous absorption), the aerosol extinction as well as the continuous absorption is found to have a negative temperature dependence. When considering the water vapor absorption continuum we need to estimate not only the value of the aerosol extinction coefficients but also their temperature dependence.

For estimating the temperature dependence of aerosol extinction we use experimental data obtained when studying the water vapor continuum in the laboratory cell and published in Ref. 1 where the dependences are presented of laser radiation transmission at two wavelengths, 0.44 and 0.63 μm, on the air relative humidity. In Ref. 1 it is shown that the radiation extinction at these wavelengths is fully due to the scattering by water-coated submicron aerosol, therefore for estimating the aerosol absorption coefficients in the spectral range from 8 to 12 μm one can use the Rayleigh formula[2]

$$\alpha_{\mathrm{a}}^{\mathrm{ab}}(\lambda) = \frac{V}{\lambda}\,\frac{36\pi\, m\, \varkappa}{(m^2 - \varkappa^2 + 2)^2 + 4\, m^2\, \varkappa^2},$$

where m and $\varkappa$ are the real and imaginary parts of the complex refractive index of submicron aerosol particles; V is the specific volume of the aerosol matter (space factor). Optical constants of aerosol particles were calculated by the mixture rules, as well as in Ref. 3:

$$m = (m_{\mathrm{d}} - m_{\mathrm{w}})(V_{\mathrm{d}}/V) + m_{\mathrm{w}},$$

$$\varkappa = (\varkappa_{\mathrm{d}} - \varkappa_{\mathrm{w}})(V_{\mathrm{d}}/V) + \varkappa_{\mathrm{w}},$$

where m_{d}, $\varkappa_{\mathrm{d}}$, V_{d}, m_{w}, $\varkappa_{\mathrm{w}}$, V_{w} are the optical constants and the specific volume of dry aerosol matter and water, respectively; $V = V_{\mathrm{d}} + V_{\mathrm{w}}$ is the specific volume of aerosol particles.

The aerosol absorption coefficients were estimated for the wavelength of 10.6 μm. The values of optical constants of dry aerosol matter and water,[4] $\alpha_{\mathrm{a}}^{\mathrm{ab}}(10.6)$, for the three versions of calculation are given in Table I. The first version of calculation corresponds to the pure water absorption, while in the second version the optical constants for dry aerosol matter are taken from Ref. 5, for the third version m_{d} and $\varkappa_{\mathrm{d}} = 1.5$. The space factors V_{d} and V_{w} were calculated by a single-parameter model[6] where the input parameter is the aerosol extinction coefficient at the wavelength of 0.55 μm.

TABLE I. Optical constants of dry aerosol matter and water in the 10.6 μm *range.*[4]

N	m_{d}	$\varkappa_{\mathrm{d}}$	m_{w}	$\varkappa_{\mathrm{w}}$
1	1.143	0.07	1.143	0.07
2	1.5	0.7	1.143	0.07
3	1.5	1.5	1.143	0.07

It should be noted that the formulas for calculating V_{d} and V_{w} were obtained for real atmospheric conditions, therefore the confidence criteria for the calculated aerosol absorption coefficients were not the values of optical constants of dry aerosol matter but the results of a comparison of $\alpha_{\mathrm{a}}^{\mathrm{ab}}(10.6)$ with the

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estimates of the cell aerosol contribution to the radiation extinction at 10.6 μm from Refs. 1, 7, and 8. As the initial material for calculating the coefficient α(0.55), we used the data on the radiation transmission at the wavelengths of 0.44 μm and 0.63 μm (Ref. 1). The Angström formulas[9] were used for the interpolation of aerosol extinction to the range of 0.55 μm. The values of the aerosol extinction coefficients α(0.55) obtained and relative air humidities, *r*, corresponding to the above coefficients are given in Table II.

TABLE II. The aerosol extinction coefficients at the wavelength of 0.55 μm *and the corresponding relative humidities of air in the laboratory cell.*[1]

α(0.55), km^{-1}	0.02	0.03	0.05	0.07	0.13	0.18
r, %	30	45	60	75	90	95

The results of calculations of the aerosol absorption coefficient by submicron aerosol for the region 10.6 μm, depending on the air relative humidity, are shown in Fig. 1. With the increase in *r* from 30 to 95%, the coefficients $\alpha_a^{ab}(10.6)$ varied within the following limits: for the first version of calculations from 0.0006 to 0.0027 km^{-1}; for the second version from 0.0040 to 0.0139 km^{-1}; for the third version from 0.0073 to 0.0281 km^{-1}.

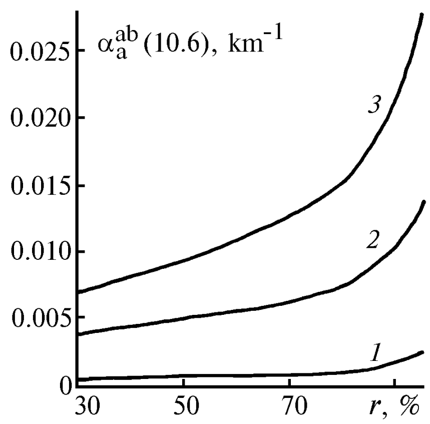

FIG. 1. Dependence of the submicron aerosol absorption coefficient in the 10.6 μm *range on the relative air humidity for three versions of* $\alpha_a^{ab}(10.6)$ *calculations* ($m_d = 1.143$, $\varkappa_d = 0.07$ *(1);* $m_d = 1.5$, $\varkappa_d = 0.7$ *(2);* $m_d = 1.5$, $\varkappa_d = 1.5$ *(3)).*

In Refs. 1, 7, and 8 the problem is discussed on the interference of aerosol with the water vapor absorption continuum in the laboratory cell. As was pointed out in Ref. 7, at relative humidities $r < 75–80\%$ ($t = 20$ °C, the absolute humidity $a < 13–14$ g/m^3), the contribution of water-coated aerosol to the laser radiation absorption at 10.6 μm in the cell experiments even in the limiting case did not exceed 12–15%. Table III presents the coefficients of aerosol $\alpha_a^{ab}(10.6)$ and continuum $\alpha_c(10.6)$ absorption,[10] and their relations, expressed in per cent, for the three versions of calculations of $\alpha_a^{ab}(10.6)$, two values of α and four different relative humidities *r* (the air temperature *t* is given in parenthesis). It can be seen from the table that for $r < 80\%$ ($t = 20$ °C, $a < 14$ g/m^3), the calculated values of $\alpha_a^{ab}(10.6)/\alpha_c(10.6)\cdot 100\%$ for the first version of the calculation do not exceed 0.6%, for the second one – 4% and for the third one – 7%. Thus, even in the limiting case the value of $\alpha_a^{ab}(10.6)$ turned out to be approximately two times smaller than the estimate[7] of the contribution of the cell aerosol to the radiation extinction (10.6 μm).

Reference 1, for making an additional estimate of a possible aerosol contribution to the radiation extinction (10.6 μm), presents spectral behavior of the extinction coefficients as well as of the scattering and absorption coefficients, calculated by Mie theory for $r = 50\%$ and the visibility of 50 km taken from Ref. 11. In the calculations of Ref. 11, the three-mode lognormal particle size distribution with the radii of 0.0085, 0.036, and 0.51 μm at the particle number densities of $5\cdot 10^4$, $28\cdot 10^3$ and 4 particles in 1 cm^3, respectively, was used. Under the above conditions, the aerosol extinction coefficient in the 10.6 μm region proved to be eCual to ~ 0.02 km^{-1}. In the laboratory cell, the visibility of 50 km ($\alpha(0.55) \approx 0.07$ km^{-1}) corresponds to the relative humidity of 75, but not 50%, as in Ref. 11, and in this case the value of $\alpha_a^{ab}(10.6)$ for the third version is 0.014 km^{-1}. Thus, the calculated values of $\alpha_a^{ab}(10.6)$ do not exceed the estimates of aerosol extinction in the laboratory cell, performed in Refs. 1, 7, and 8.

Let us now consider the dependence of aerosol extinction on the relative air humidity and the effect of the neglect of this dependence on the estimate of temperature dependence of water vapor absorption continuum. In Ref. 1 it was emphasized that the experimental data, determining the dependence of transmission at 0.44 and 0.63 μm on the relative humidity, are practically on one curve at different ambient temperature. Taking this into account, we constructed the dependences of $\alpha_a^{ab}(10.6)$ on the absolute humidity at different temperatures. Figure 2 presents such dependences for $m_d = 1.5$, $\varkappa_d = 0.7$ and five values of air temperatures. This figure shows that aerosol extinction has the negative temperature dependence, namely, $\alpha_a^{ab}(10.6)$ increases with the decrease of air temperature (at a constant absolute humidity). Thus the presence of aerosol in the laboratory cell, when investigating the water vapor absorption continuum, will result in overestimation of its negative temperature dependence.

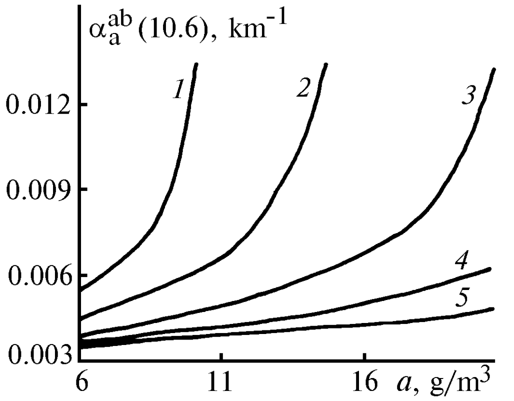

FIG. 2. Dependence of the submicron aerosol absorption coefficient in the 10.6 μm *range on the absolute air humidity for* $m_d = 1.5$, $\varkappa_d = 0.7$ *and five values of air temperature* $t = 12$, 18, 24, 30, 36°C *(curves 1, 2, 3, 4, 5, respectively).*

For assessing possible overestimation of the temperature dependence of continuous absorption, Table IV presents the temperature coefficients of aerosol $K_a = [\alpha_a^{ab}(r_2) - \alpha_a^{ab}(r_1)]/(t_2 - t_1)$ and continuous $K_c = [\alpha_c(r_2) - \alpha_c(r_1)]/(t_2 - t_1)$ (Ref. 10) absorption by water vapor and their ratios $K_a/K_c \cdot 100\%$ for different ranges of relative air humidity r_1–r_2 (the corresponding temperature ranges t_1–t_2 are given in parenthesis), two values of the absolute humidity a and three versions of calculations of $\alpha_a^{ab}(10.6)$. The contribution of aerosol to the estimate of temperature dependence of water vapor absorption continuum grows with the increase of relative air humidity and decreases with the increase of absolute humidity. Tables III and IV give the evidence that the submicron aerosol[1] distorts the estimate of the continuum temperature dependence by approximately two to ten times more strongly than the value of estimated coefficients of continuous absorption $\alpha_c(10.6)$. This is explained by the fact that the temperature coefficient of aerosol absorption K_a, being the first derivative of $\alpha_a^{ab}(10.6)$ with respect to the ambient temperature (at the fixed absolute air humidity), varies in t much faster than the coefficient $\alpha_a^{ab}(10.6)$. So, for $a = 10$ g/m^3, $\varkappa_d = 0.7$ and when passing from the humidity range of 33–46% to 78–94% the value of K_a increases by a factor of 19, while for r change from 33% to 94% $\alpha_a^{ab}(10.6)$ increases only by a factor of 3.2. In this case the values of the coefficients K_c and $\alpha_c(10.6)$ vary by a factor of 1.6 and 1.3, respectively. As a result, the cell aerosol strongly affects the estimate of the temperature dependence of the continuous absorption coefficients. At $a = 10$ g/m^3 and for the range r_1–r_2 = 33–94% the neglect of the submicron aerosol extinction with $\varkappa_d = 0.07$, 0.7 and 1.5 will result in an increase of the estimates of the continuum temperature dependence by 5%, 25% and 51%, respectively.

TABLE III. Coefficients of aerosol, $\alpha_a^{ab}(10.6)$*, and water vapor continuous,* $\alpha_c(10.6)$, (Ref. 10) *absorption and their ratios* $\alpha_a^{ab}(10.6)/\alpha_c(10.6) \cdot 100\%$ *for different relative humidities of air, r (corresponding temperatures t are given in parenthesis).*

r, % (t, °C)	$\alpha_c(10.6)$. km^{-1}	$\varkappa_d = 0.07$		$\varkappa_c = 0.7$		$\varkappa_d = 1.5$	
		$\alpha_a^{ab}(10.6)$, km^{-1}	$\alpha_a^{ab}/\alpha_c \cdot 100\%$	$\alpha_a^{ab}(10.6)$, km^{-1}	$\alpha_a^{ab}/\alpha_c \cdot 100\%$	$\alpha_a^{ab}(10.6)$, km^{-1}	$\alpha_a^{ab}/\alpha_c \cdot 100\%$
				$a = 10$ g/m^3			
33(30)	0.1095	0.0006	0.5	0.0041	4	0.0076	7
46(24)	0.1196	0.0007	0.6	0.0047	4	0.0090	8
65(18)	0.1319	0.0010	0.8	0.0061	5	0.0118	9
78(15)	0.1391	0.0012	0.9	0.0075	5	0.0147	11
94(12)	0.1470	0.0025	1.7	0.0133	9	0.0269	18
				$a = 14$ g/m^3			
33(36)	0.1786	0.0006	0.3	0.0041	2	0.0076	4
46(30)	0.1954	0.0007	0.4	0.0047	2	0.0090	5
64(24)	0.2158	0.0010	0.5	0.0060	3	0.0117	5
76(21)	0.2275	0.0012	0.5	0.0072	3	0.0140	6
80(20)	0.2317	0.0015	0.6	0.0083	4	0.0164	7
91(18)	0.2405	0.0022	0.9	0.0114	5	0.0229	10

TABLE IV. Temperature coefficients of aerosol, K_a, and water vapor continuous, K_c, (Ref. 10) absorption and their ratios $K_a/K_c \cdot 100\%$ for different ranges of relative air humidity r_1–r_2 (corresponding temperature ranges t_1–t_2 are given in parenthesis).

r_1–r_2, % (t_1–t_2, °C)	K_c, km^{-1} deg^{-1}	$\varkappa_d$ = 0.07		$\varkappa_d$ = 0.7		$\varkappa_d$ = 1.5	
		K_a, km^{-1} deg^{-1}	$K_a/K_c \cdot 100\%$	K_a, km^{-1} deg^{-1}	$K_a/K_c \cdot 100\%$	K_a, km^{-1} deg^{-1}	$K_a/K_c \cdot 100\%$
				a = 10 g/m^3			
33–46 (30–24)	– 0.00168	– 0.00002	1	– 0.00010	6	– 0.00023	14
46–65 (24–18)	– 0.00205	– 0.00004	2	– 0.00023	11	– 0.00047	23
65–78 (18–15)	– 0.00240	– 0.00009	4	– 0.00047	20	– 0.00097	40
78–94 (15–12)	– 0.00263	– 0.00042	16	– 0.00193	73	– 0.00407	155
33–94 (30–12)	– 0.00208	– 0.00011	5	0.00051	25	– 0.00107	51
				a = 14 g/m^3			
33–46 (36–30)	– 0.00260	– 0.00002	1	– 0.00010	4	– 0.00023	9
46–64 (30–24)	– 0.00340	– 0.00004	1	– 0.00022	6	– 0.00045	13
64–76 (24–21)	– 0.00390	– 0.00007	2	– 0.00040	10	– 0.00077	20
76–91 (21–18)	– 0.00433	– 0.00036	8	– 0.00140	32	– 0.00297	69
33–91 (36–18)	– 0.00344	– 0.00009	3	– 0.00041	12	– 0.00085	25

Thus, when investigating experimentally the temperature dependence of water vapor absorption continuum in the spectral range of 8–12 μm, we should carefully study the problem on exclusion of even a small aerosol extinction depending on the relative air humidity.

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