

## IR—SPECTROSCOPIC STUDIES OF ATMOSPHERIC ABSORPTION CHARACTERISTICS (LITERATURE REVIEW)

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*The results of various IR-spectroscopic studies of the atmospheric aerosol refractive index ( $\kappa$ ) are analyzed. Minimum and maximum estimates of  $\kappa$  are obtained for urban and rural aerosols. A comparison is made of the experimental and model  $\kappa$  values in the spectral range from 3 to 20  $\mu\text{m}$ .*

### CLASSIFICATION AND MAIN AEROSOL ABSORPTION BANDS IN VARIOUS REGIONS

Aerosol, is the most unstable atmospheric component both in space and time. At the present time there is no unique approach to aerosol classification.

At the same time the classification given in Ref. 1 and presented below as well as in Table I, in our opinion, reflects the actual variety of aerosol types: 1) dust-like, 2) water-soluble, 3) soot, 4) oceanic, sea salt, 5) mineral, crystalline, 6) sulfate, and 7) biological.

TABLE I

*Types of tropospheric aerosols.<sup>1</sup>*

Aerosol types	Components <sup>*)</sup>	Region
(A) Pure continental, continental background	2, 1	Most of Africa south of the Sahara, South America, Asia
(B) Pure forest	2,7,1	thick forests of the Amazon, Asia, and Africa
(C) Average continental	2,3,1	Regions slightly influenced by human pollution
(D) Rural	2,1,7	Rural environment
(E) Mineral (pure desert)	5	Arid and semi-arid environment
(G) Pure marine	4,2	Southeast Pacific, south equatorial Atlantic, Indian Ocean
(H) Mixture of marine and mineral	4,2,5	North equatorial Atlantic (trade-wind region)
(I) Marine (polluted areas of the sea)	4,2,3	Mediterranean, North Atlantic
(J) Pure polar	4, 6	Antarctic
(K) Polar	3,5,4	Arctic
(L) Urban, industrial	2,3,1	Industrialized regions

\*) The key to the classification of aerosol components is given in the text.

We must however note that the terminology used in Ref. 1 needs some additional clarification. In particular, it refers to an oceanic aerosol composition as well as to the difference between dust-like and mineral (crystalline) components. For example, besides sea salt the oceanic aerosol composition must include a sulfate fraction because only in this case is the classification proposed in Ref. 1 in good agreement with the well-known facts of sulfate presence in the arctic aerosol.<sup>2,3</sup>

The analysis of the IR spectra of aerosols over different regions shows that the aerosol spectra of practically all above-mentioned types, except pure desert and sea aerosol, reveal general features in the IR region (3–20  $\mu\text{m}$ ). They contain, as a rule, not more than 5 quite intense absorption bands centered near 3, 6, 7, 9, 10, and 16  $\mu\text{m}$  (Table II) with relative band intensities which vary due to the concentration of the individual components in the aerosol sample.

This is connected with both the spectral features of the aerosol particles and the remote transfer of a submicron aerosol fraction and also of the gaseous pollutants which take part in aerosol formation. These factors lead to an equalizing of the aerosol chemical composition all over the globe.

TABLE II

*The main absorption bands of atmospheric aerosols from various regions.*

Spectral range		Chemical groups mainly contributing to absorption		Comments
$\lambda, \mu\text{m}$	$\nu, \text{cm}^{-1}$	Inorganic	Organic	
2.8-3.5	3500-2850	$\text{NH}_4^+, \text{OH}^-$	$\text{CH}_3, \text{CH}_2$	Narrow organic bands over the inorganic wide band contour
5.7-6.25	1750-1600	$\text{OH}^-$	$\text{C=O}, \text{C=C}, \text{OH}^-$	Organic absorption prevails
6.7-7.5	1500-1330	$\text{NH}_4^+, \text{CO}_3^{2-}, \text{NO}_3^-$	$\text{CH}_3, \text{CH}_2$	Inorganic absorption prevails
8.3-11	1200-910	$\text{SO}_4^{2-}, \text{SiO}_2, \text{SiO}_4^{4-}$	$\text{C-O}, \text{C-C}$	Inorganic absorption prevails
16.1-16.4	620-610	$\text{SO}_4^{2-}$	—	—

**CRITICAL ANALYSIS OF SOME METHODS FOR DEFINING THE IMAGINARY PART OF THE COMPLEX REFRACTIVE INDEX  $M = n - i\kappa$  OF THE ATMOSPHERIC AEROSOL**

Two methods may be used to obtain  $\kappa$  of a real aerosol: 1) measure the absorption of the individual aerosol components and then calculate the total values of  $\kappa$ ; 2) measure the absorption of the aerosol samples themselves. The following expression for the

transparency  $T$  is basic for calculating  $\kappa$ :

$$T_\lambda = \exp(-\alpha_\lambda l) = \exp(-4\pi\kappa_\lambda l/\lambda); \quad (1)$$

$$\alpha_\lambda = 4\pi\kappa_\lambda/\lambda, \quad (2)$$

where  $\alpha_\lambda$  is the spectral absorption coefficient (SAC) per unit layer depth  $\partial$ ;  $\kappa_\lambda, \text{g}^{-1} \cdot \text{cm}^2$  is SAC per unit surface density of the aerosol sample  $M$ .

This expression is not accurate for a real aerosol sample or for a dispersed single-component medium, and therefore measurements of this type contain a number of systematic errors.

1. Uncertainty in the interpretation of  $\kappa$  in the case of an aerosol sample. For example, for  $\kappa = 0.05$  the following variants are possible: the total aerosol mass has  $\kappa = 0.05$  or 10% of it has  $\kappa = 0.5$ , and 90% —  $\kappa = 0$ , etc. The single scattering albedo of the aerosol in the atmosphere is different for different cases. Therefore when investigating real aerosol samples, knowledge of the particle size distribution is desirable: rough dispersed (RD)  $r > 1 \mu\text{m}$ , submicron (SM)  $0.1 < r < 1 \mu\text{m}$  and microdispersed (MD)  $r < 1 \mu\text{m}$ , with different chemical compositions due to different origin ( $r$  is the effective radius of the aerosol particles). In addition, real aerosol samples must be divided into their soluble and insoluble components.

2. The absence of the scattering assumed by expression (2) cannot always be realized in the case of disperse media. A numerical experiment<sup>4</sup> has shown that if the scattering is neglected, it leads to measured values which overestimate the real ones in the spectral range of weak absorption (the band wings).

3. The applicability of Eq. (2) is limited for aerosol particles (deviation from linearity). Formula (2) gives a good approximation for the particles with  $r_d \leq 0.1\lambda$ , a quite narrow particle size distribution  $\sigma_d \leq 1$ , not very large values of  $n$ , and  $\kappa \leq 1$  (Refs. 4–7). The numerical experiment<sup>4</sup> showed that in the disperse medium the deviation from linearity, when  $\alpha_\lambda$  depends on  $\kappa_\lambda$ , leads to measured  $\kappa_\lambda$  values which underestimate the real ones at the band maxima.

*The KBr tableting method* has been developed and widely used by Volz.<sup>8–11</sup> The essence of this method is that it minimizes scattering losses by choosing an immersion medium with refractive index close to that of the aerosol. The value of  $\kappa$  is determined by measuring the transparency of the aerosol sample, milled and pressed into the KBr-tablet, using the formula

$$\ln(T_\lambda/T_{\lambda a}) = K_\lambda M = \frac{4\pi\kappa}{\rho l}, \quad (3)$$

where  $T_\lambda$  and  $T_{\lambda a}$  are the transparency values of pure tablet and the aerosol tablet, respectively, and  $\rho$  is the aerosol particle density.

The causes of the deviation of dependences (2) and (3) from linearity (see paragraph 3 above) are, to a great extent, reduced after repeated milling.<sup>9</sup>

Volz<sup>9</sup> had obtained approximately a twofold increase of  $\kappa$  in comparison with a single milling. Thus, the results obtained using the KBr-tablet method in the single milling of the sample must be corrected.

The thin film method has been developed and used by K. Fischer<sup>12-14</sup> for SM aerosol investigation. A moving substrate is covered by a thin layer  $l$  of the aerosol sample. The value of  $\kappa_{\lambda f}$  the aerosol film so obtained is given by the expressions

$$\kappa_{\lambda r} = \frac{\lambda}{4\pi\Delta l} \ln [I_{\lambda}(l)/I_{\lambda}(l + \Delta l)]; \quad (4)$$

$$\kappa_{\lambda r} \rho = \lambda [4\pi (m/S - m'/S')]^{-1} \ln I_{\lambda}/I_{\lambda}, \quad (4')$$

where  $\Delta l$  is the difference of the thicknesses of two thin aerosol layers evaporated on the substrate sequentially,  $m$  and  $S$  are the mass and surface area of the sample, and  $\rho$  is the density of the aerosol. Fischer<sup>14</sup> derived expressions for the absorption coefficient of the aerosol particles themselves  $\kappa_{\lambda a}$  with the help of the theory of continuous media and variance analysis. These values of the aerosol absorption coefficient,  $\kappa_a$ , seem to be roughly 30% higher than the corresponding values of  $\kappa_f$ . According to the data of Ref. 4, the error in determining  $\kappa$  is  $\delta\kappa/\kappa \sim \delta l/l$ . Thus the condition  $\Delta l \ll l$  gives good accuracy. This condition is not satisfied in Ref. 15, where  $\Delta l \sim l$ , which results in a 100% error in measuring  $\kappa$ .

Taking the data from Refs. 6-7 into account, Fischer assumes expression (2) to hold with high accuracy for his experiments ( $\lambda = 2.5-17 \mu\text{m}$  for particles with  $r < 1$  and  $\kappa \leq 1.2$ ). In our opinion, this is not the case for the spectral range  $\lambda = 2.5-6 \mu\text{m}$ , where scattering cannot be neglected. Moreover, for the case of an insufficiently narrow size distribution  $\sigma_d \geq 1.5-2$ , which Fischer does not allow for,  $\kappa_{\lambda}$  depends on the particles radius and  $\sigma_d$ , which leads to a decrease in  $\kappa_{meas}$  as compared to  $\kappa_{real}$ .<sup>5</sup> Hence the  $\kappa$  values at the band maxima in Fischer's measurements may be too low.

Figure 1 shows the finalized results of spectral behavior measurements of crystalline, ground, and urban aerosols, where the  $\kappa_a$  values at the band maximum near  $9.5 \mu\text{m}$ , obtained by Fischer, are close to their minimal values. A more extended comparison of Fischer's and Volz's data also shows that the  $\kappa$  values obtained by Fischer are 2-3 times smaller than those obtained by Volz.

The monolayer method was used by Lubovtseva and Gabelko<sup>21, 23</sup> to determine the aerosol spectral absorption coefficient  $\alpha_{\lambda}$ ,  $\text{km}^{-1}$  in situ. The method assumes nonoverlapping of the aerosol particles evaporated on the substrate. According to Cabelko<sup>23</sup> for the overlapping coefficient  $2 \geq \eta \geq 0.6$  the error is equal to 40-80%, and for  $\eta > 2$  the method is no longer valid. Using the monolayer approximation

$$\alpha_{\lambda} = [1 - I/I_0(\lambda)] \frac{S}{QtW}, \quad (5)$$

where  $S$  is the cross-sectional area of the light beam,  $Q$  and  $t$  are the flow rate and time of air pumping through the impactor, and  $W$  is the impactor entrapment coefficient. Our analysis of the sampling conditions in Refs. 21 and 22 shows that the necessary values of the overlapping coefficients are not guaranteed ( $\eta = 0.1-9$ ), and the experimental data on aerosol monolayer absorption obtained with the help of the impactor<sup>21, 22</sup> and recorded by an ordinary IR-spectrometer (IKS-29) do not exceed the noise level. Hence the  $\alpha_{\lambda}$  data<sup>21, 22</sup> should be taken into account but carefully.

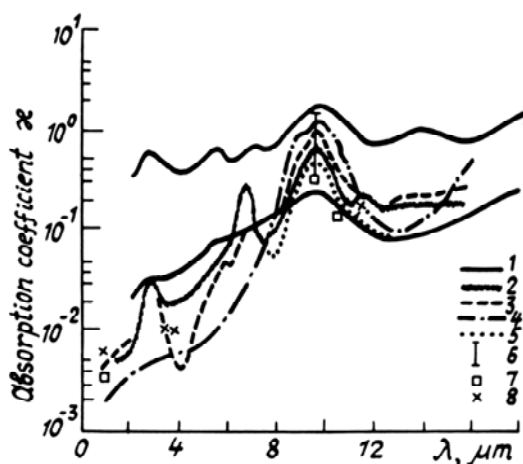


FIG. 1. The absorption coefficient of crystalline aerosols with a predominance of argillaceous minerals and also of urban aerosols. 1 - dust aerosol, the city of Bedford<sup>9</sup>; 2 - ground aerosol<sup>16</sup>; 3 - Sahara dust from the island of Barbados<sup>8</sup>; 4 - basalt, reflection from a massive sample<sup>17</sup>; 5 - SM arid aerosol, the thin film method<sup>12</sup>; 6 - ground aerosol<sup>18</sup>; 7, 8 - ground aerosol, the spectrophone method<sup>19-20</sup>; 1-3 and 6 - the KBr-technique.

Transformation of  $\alpha_{\lambda}$  into  $\kappa_{\lambda}$  as well as  $\kappa_{\lambda}$  into  $\alpha_{\lambda}$  was made in Refs. 15. 21-27 using Hänel's formula:<sup>28</sup>

$$\alpha_{\lambda} = \frac{12\pi}{\lambda} \frac{\kappa}{\rho} \frac{1}{n^2 + 2} \frac{M}{V}, \quad (6)$$

where  $M/V$  is aerosol particle mass per unit volume of air. Formula (6) is valid for the spectral ranges  $\lambda = 0.55-2 \mu\text{m}$  and  $9.25-12 \mu\text{m}$ , and  $\kappa \leq 1$  (Ref. 2). Significant errors may arise due to errors in the formula for determining the mass of the absorbing component if the latter is small compared to the total aerosol mass. This took place, for example, when  $\alpha_{org}$  of the organic component was determined from  $\kappa_{org}$ .

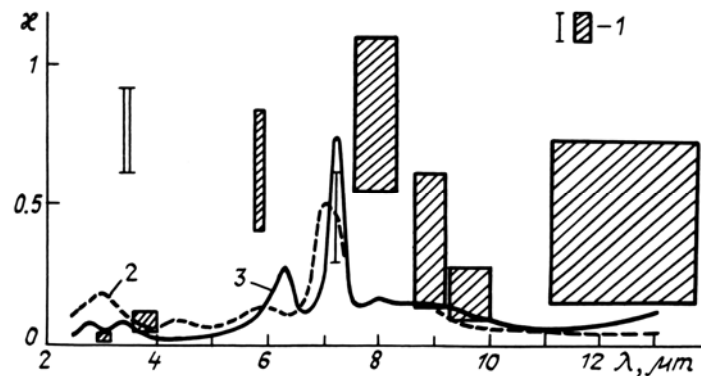


FIG. 2. The spectral dependence of the absorption coefficient of the aerosol organic component. 1 –  $\kappa_{org}$ , Pasadena, California<sup>21,25,26</sup>; 2, 3, –  $\kappa_{org}$ , Mainz, FRG.<sup>9</sup>

The values of  $\kappa_{org}$  (Refs. 21, 24, and 25) were taken from a literature review<sup>29</sup> of data on molar absorption coefficients  $\varepsilon_{\lambda a}$  of different characteristic groups of organic compounds. The values  $\varepsilon_{\lambda a}$  are “rough estimates”.<sup>29</sup> Hence, according to the results of our data analysis<sup>21, 24, 25, 29</sup> the values of  $\kappa_{org}$  (Refs. 21, 24, and 25) seem to be only approximate. The rather wide spectral intervals in which the  $\kappa_{org}$  values are given, in view of the fact that the  $\varepsilon_{\lambda a}$  values are referred to the absorption band maxima of the corresponding characteristic groups, and significant variations of the  $\kappa_{org}$  values themselves confirm our argument (see Fig. 2).

The values  $\kappa^{RU}$  (Refs. 15, 21, 22, 24–27) for the rural and urban environments were calculated taking into account the principal absorbing components of the SM fraction: ammonium sulfate (AS), soot (S), and organic material (O) by the formula

$$\kappa_{sm} = \kappa_{org} a_{org} + \kappa_{as} a_{as} + \kappa_s a_s, \quad (7)$$

where  $\kappa_{as}$  is taken from Ref. 4 and  $\kappa_s$  from Ref. 30. and  $a_i = \frac{m_i}{m_{sm}}$  is the relative mass concentration of the  $i$ -th component measured experimentally.

Analysis of the data<sup>21, 25, 26</sup> shows that the influence of the organic component on  $\kappa_{sm}$  due to small values of the relative concentrations of the organic material are not significant even in those spectral intervals where  $\kappa_{org}$  exceeds or is comparable with  $\kappa_{as}$  and/or  $\kappa_s$ . It is seen from a comparison of the data (Figs. 2 and 3) that even for significant variations of  $\kappa_{org}$  the variations of  $\kappa_{sm}$  are small.<sup>21, 25, 26</sup>

The  $\kappa_{RD}$  values used to calculate  $\kappa_{\Sigma} = \kappa_{sm} \frac{m_{sm}}{m_{\Sigma}} + \kappa_{RD} \frac{m_{RD}}{m_{\Sigma}}$  in Refs. 21, 24–27 were taken from Ref. 16. With the most typical, in the opinion of the authors of Refs. 21, 24–27, mass ratio  $m_{sm} : m_{RD} = 2 : 3$ , the  $\kappa_{\Sigma}$  values in the majority of the spectral intervals are at least twice as small as  $\kappa_{sm}$ . This means that the contribution of  $\kappa_{RD}$  to  $\kappa_{\Sigma}$  can be neglected.

On the basis of a comparison of the results given by different authors we may take the  $\kappa_{sm}^{RU}$  values given by Lubovtseva and Cabelko as an upper estimate and the Fischer's  $\kappa_{sm}$  values as a lower estimate for  $\kappa_{sm}$  of continental aerosols.

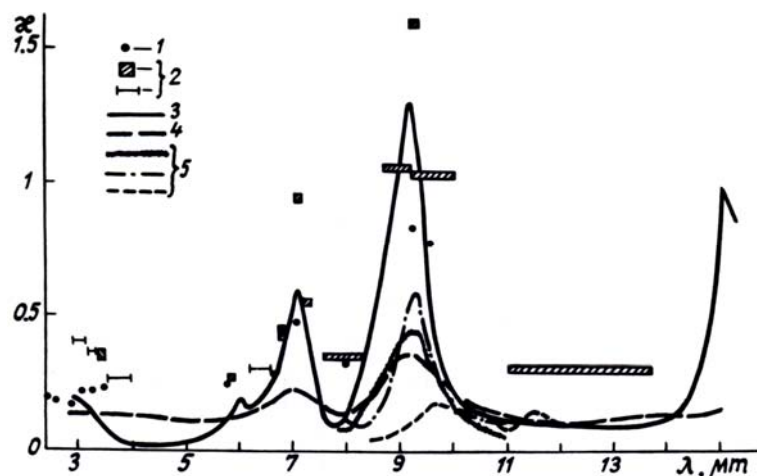


FIG. 3. The spectral dependence of the absorption coefficient of urban SM aerosol  $\kappa_{sm}^{urb}$ , 1 – Moscow,<sup>15</sup> the thin film method<sup>12</sup>; 2 – Pasadena<sup>21</sup> calculated by Eq. (7); 3 – Mappen, the KBr-technique<sup>9</sup>; 4 – Mainz, experiment<sup>12</sup>; 5 – Mainz, experiment,<sup>14</sup> the method of Ref. 12 plus variance analysis.<sup>12</sup>

**COMPARISON OF EXPERIMENTAL VALUES OF  $\kappa$  WITH SOME AEROSOL MODELS**

In constructing aerosol models it is important to take adequate account of the role of individual aerosol components. Ivlev and Popova in their well-known model<sup>31</sup> considered only the mineral component. G. V. Rozenberg,<sup>32-34</sup> in his time, objected to this approach. However, the investigations of Andreev and Ivlev<sup>35-37</sup> and Lubovtseva and Gabelko showed that the role of the organic material in absorption by the submicron continental aerosol was not determined.

Figure 4 presents a comparison of the values of  $\alpha_{org}^{R,U}$  according to the data given by Ivlev-Andreev and Lubovtseva-Gabelko with  $\alpha_{min}^U$  (Ref. 31). It can be seen that the mineral component absorption prevails over the entire spectral range over the organic one for urban and rural aerosols.

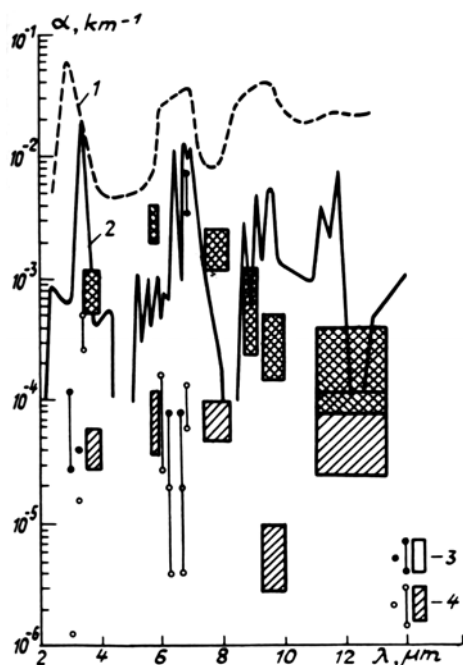


FIG. 4. The spectral absorption coefficients  $\alpha$ ,  $km^{-1}$ , of the organic and mineral components of urban and rural aerosol; 1 -  $\alpha_{min}^{urb}$  (Ref. 31); 2 -  $\alpha_{org}^{urb}$  (Refs. 36 and 37); 3 -  $\alpha_{org}^{urb}$  (Refs. 21 and 25); 4 -  $\alpha_{org}^{rur}$  (Refs. 21 and 25).

Figure 5 presents a comparison of  $\kappa_{min}^{mod}$  values<sup>31</sup> with a number of  $\kappa_a^{exp}$  values. The notable excess of the  $\kappa_{sm}^{exp}$  values over the model ones in the regions 3-4  $\mu m$  and 5.5-6.5  $\mu m$  cannot be explained entirely by the fact that the organic component is not taken into account<sup>31</sup> if  $\kappa_{org}$  and  $m_{org}$  data<sup>21, 25</sup> are used.

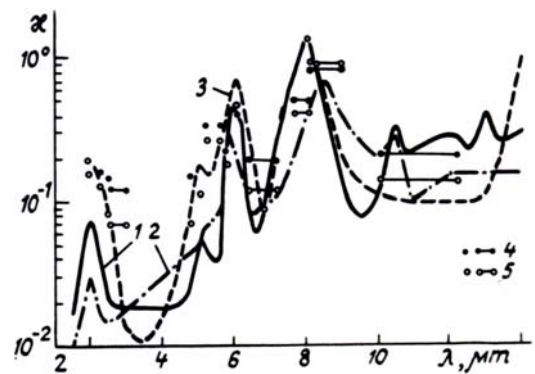


FIG. 5. The spectral dependences of the absorption coefficients of different aerosol types: 1 -  $\kappa_{min}^{calc}$  (Ref. 31); 2-5 - experiment: 2 -  $\kappa_{cd}$  is ground aerosol (Ref. 16); 3 -  $\kappa_{sm}^{urb}$  is the water soluble fraction (Ref. 9); 4, 5 -  $\kappa_{\Sigma}^{urb}$ ,  $\kappa_{\Sigma}^{rur}$  (Refs. 21 and 25).

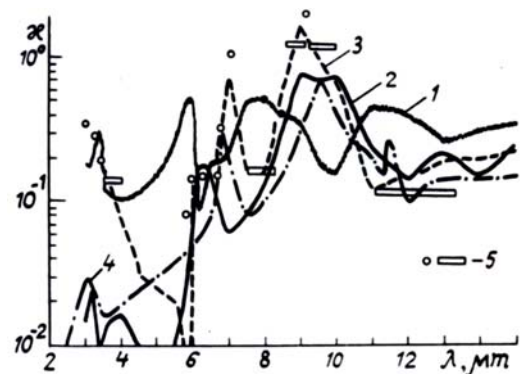


FIG. 6. The spectral behavior of the absorption coefficient  $\kappa$  of the rural aerosol according to the following data: Ref. 38 - 1 (RD); 2 (SM); 3 (MD); Ref. 16 - 4 (RD); Refs. 21, 25 - 5 (SM).

In the development of their model,<sup>31</sup> Andreev and Ivlev suggested empirical models for  $\kappa_{\lambda}$  of the MD, SM, and RD aerosol fractions.<sup>38</sup> Figure 6 shows a comparison of these data with  $\kappa_{sm}^{exp}$  (Refs. 21 and 25) and  $\kappa_{RD}^{exp}$  (Ref. 16). As can be seen from the figure,  $\kappa_{sm}^{mod}$  and  $\kappa_{md}^{mod}$  (Ref. 38) provide a good description of  $\kappa_{rd}^{exp}$  (Ref. 16) and  $\kappa_{sm}^{exp}$  (Refs. 21, 25), respectively, while  $\kappa_{rd}^{mod}$  does not describe the real RD aerosol.

K.Ya. Kondrat'ev and N.I. Moskalenko<sup>2</sup> give model values of  $\kappa$  for the water-soluble and -insoluble aerosol fractions which practically coincide with the analogous values of  $\kappa$  from Ref. 39. Figure 7 presents a comparison of  $\kappa$  (Refs. 2, 39) with Volz's experimental data<sup>9</sup> of  $\kappa_{sm}^{sol}$  and with  $\kappa_{RD}$  of E.M. Patterson.<sup>16</sup> The latter refer to the ground

aerosol and consist mainly of insoluble components. Figure 7 shows that the experimental  $\kappa$  values exceed the corresponding model ones (by from 1.5 to 10 times) over practically the entire spectral range. The regions near 4.5  $\mu\text{m}$  for soluble component and near 12  $\mu\text{m}$  for the Insoluble one are exceptions.

Thus, the Ivlev-Popova<sup>31</sup> model is the most convenient for describing the absorption coefficient of real continental aerosol (urban and rural), taking into account the measurement errors of the  $\kappa$  values and variations of the chemical composition of the different aerosol fractions. At the same time we must bear in mind<sup>4</sup> that the approach in which first the optical constants are averaged and then the optical characteristics are derived (the single scattering albedo, the phase scattering function, the effective extinction cross section, absorption, etc.), can be used only in the case in which the optical characteristics are linear in the optical constants. In particular, for multicomponent aerosol particles,<sup>4</sup> neither the optical constants of the materials nor their optical characteristics can be averaged.

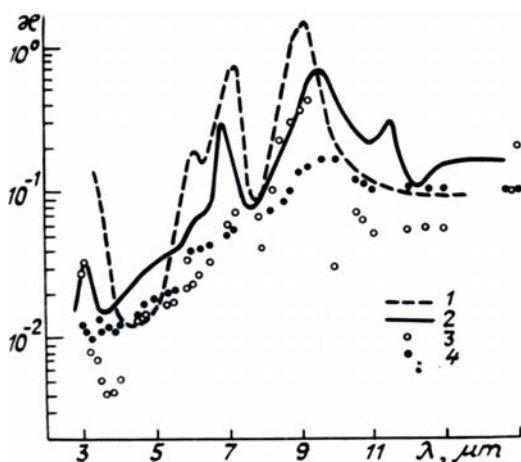


FIG. 7. Comparison of the experimental (1, 2) and model (3, 4) values of  $\kappa$  of water-soluble and -insoluble aerosol fractions: 1 – water-soluble SM-fraction of the urban aerosol,<sup>9</sup> 2 – crystalline groundaerosol;<sup>16</sup> 3, 4 – water-soluble and -insoluble fractions, respectively.<sup>2,39</sup>

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