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Size distributions of tropospheric particles in terms of the modified gamma function and relationships between skewness and mode radius

By CLAUDIO TOMASI and FRANCESCO TAMPIERI, *Sezione Microfisica dell'Atmosfera, C.N.R., Bologna, Italy*

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ABSTRACT

Size spectra of tropospheric particles measured by several authors have been interpreted using a computational method in terms of the modified gamma distribution function. The results are shown for particles of different origin: coastal and maritime, continental, desert, urban and anthropogenic.

Correlation lines have been found between the skewness of the resulting size distributions and the logarithm of the corresponding mode radius for the various types of atmospheric particles. Based on these relationships a method is proposed which is suitable for obtaining size distributions of particulate matter typical of different atmosphere.

1. Introduction

Airborne particles scatter and absorb the solar radiation and change the terrestrial radiation field in a manner depending both on their physicochemical properties and their size distributions. In order to evaluate the role played by the atmospheric particles on the thermal balance of the earth, Mie theory can be employed if both the complex refractive index and the size distribution of the particle load are known.

As pointed out by Foitzik (1965) sources of particles of different sizes can affect the same atmospheric air mass in such a way that the overall particle size distribution can present multimodal profiles. The growth and removal mechanisms such as sedimentation and coagulation can modify the original composite features of the particle size distribution and cause the mode concentrations to decrease with the radius with slopes which can be given in terms of the well-known power law of Junge (1952).

Deirmendjian (1964, 1969) proposed the modified gamma distribution function for describing particle size distributions of marine or coastal, continental and stratospheric type and for studying the light scattering phenomena. This function presents the general form:

$$n(r) = a\, r^{\alpha} \exp\left[-\frac{\alpha}{\gamma}\left(\frac{r}{r_c}\right)^{\gamma}\right], \quad 0 \le r < \infty \qquad (1)$$

where $n(r)$ is the volume concentration per unit radius of particles with radius r and the four shape-parameters a, α, γ and r_c have to be positive. The latter three parameters determine the shape of the distribution curve while the parameter a allows changing the total number N of particles per unit volume. The parameter α assumes only integer values while r_c is the mode radius.

The use of four shape-parameters which can be widely varied renders Deirmendjian's function appropriate for giving different shapes for the two wings of the same mode and for properly covering the large variability which characterizes the atmospheric particle size distributions. The modified gamma function has been utilized by Kuriyan & Sekera (1974) and by McKellar (1974) for investigating the scattering properties of the atmospheric particles. In order to obtain representative size distributions of tropospheric airborne particles an ample collection of size spectra have been interpreted by De Luisi et al. (1972) in terms of Deirmendjian's function. Their method of curve fitting does not need to involve setting a value for the mode radius r_c while the parameter α has been fixed at 1. More generally the use of Deirmendjian's function with a large range of the parameter α appears suitable for giving more appropriate representations of tropospheric particle size-distributions, especially with regard to the narrow shapes characterizing most of the size spectra of large and giant particles.

Particle size spectra measured by several authors in atmospheric air masses of different origin have been analyzed in the present paper in terms of the modified gamma distribution function. The computational procedure we have adopted determines the parameter sets α, γ, r_c which give the best-fit distribution curves.

2. Computational procedure

The computational method used for examining the empirical size spectra has been presented in a previous paper (Tampieri & Tomasi, 1976). It is based on the following stages:

(i) reduction of each size spectrum to a run of at least four pairs (r_i, n_i) where n_i is the particle concentration per unit radius (expressed in $cm^{-3}\,\mu m^{-1}$) at radius r_i (in μm);
(ii) normalization of n_i to n_1 and use of the logarithms of the obtained ratios;
(iii) search for each experimental run of the parameter set giving the best-fit solution. It is found by minimizing the function F which represents the average relative deviation of the empirical data from the distribution on logarithmic scale in the range of α from 1 to 10. The analytical form of F and the criteria adopted for selecting the good-fit solution (in the case in which the function F does not present a minimum) are given in the above-cited paper. The minimum assumed by F correspondingly to the solution has been called f_{min}.

3. Examination of the empirical data

The size spectra which show distinctly multimodal features have been divided into separate runs, each related to a single mode and independently analyzed by the computer. In spite of their probably multimodal structure most of the examined size spectra appear consistent with a unique mode. Moreover they consist of data which in general describe only the smoothed right wing. As the computational procedure does not require presetting the mode radius or the empirical data set relative to its range, the corresponding solutions which have to fit the right wing data determine also the mode radius and the shape of the smaller particle wing.

In certain cases the computational method gave solutions characterized by very low values of r_c (less than $10^{-5}\,\mu$m) and by γ less than 0.1. Correspondingly, the parameter α resulted as being most frequently equal to 1. Parameter sets of such a type give size distributions with shapes of the larger particle wing which result to be very similar to Junge type size distributions (in which the volume concentration of particles per unit radius $n(r)$ turns out to be proportional to $r^{-(\nu^*+1)}$). These empirical runs have been re-examined in terms of the power law distribution in order to evaluate the exponent ν^*. All correlation lines we have found present absolute values of the correlation coefficient very close to 1.

In the other cases, the solutions given by the computational procedure in terms of the modified gamma function will be represented as points in the three-dimensional space of the shape-parameters α, γ and r_c. With reference to the sampling sites and the remarks given by the authors, the experimental runs have been separated into the following types:

(a) *Coastal and maritime type*. We have analyzed 35 runs taken from the following authors: Kuroiwa (1956), Pueschel & Noll (1967), Blifford (1970), Harris (1971), Alkezweeny & Slinn (1972). Five solutions result of the Junge type with values of ν^* from 0.9 to 1.8. The parameter sets related to the modified gamma distribution function are presented in Fig. 1. About 87% of the solutions present values of f_{min} less than 0.1. An example of good-fit solution is given in Fig. 2, which shows how the computational method we use individuates the mode radius and determines the shape of the left wing of the distribution curve on the basis of the data set which involves only the upper radius range.

(b) *Continental type*. In all 37 runs have been examined from: Junge (1955), Cartwright et al. (1956), Blifford & Ringer (1969), Harris (1971),

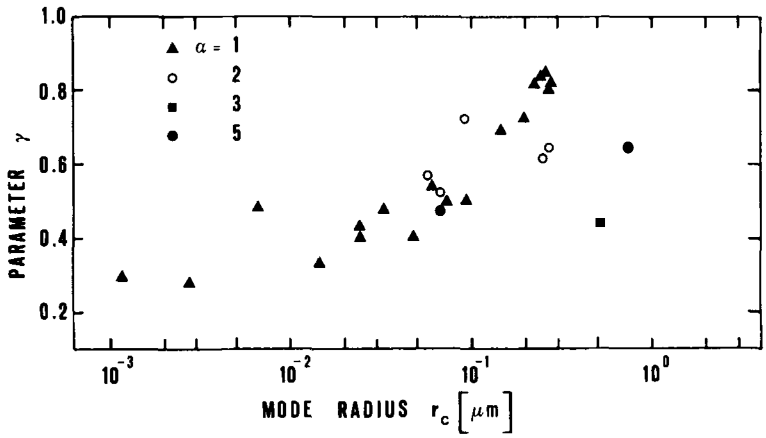

Fig. 1. Parameter sets found as fitted solutions in terms of the modified gamma size distribution for empirical size spectra of coastal and maritime type particles. The mode radius r_c is given in abscissa, the parameter γ in ordinate while the parameter α is indicated by means of different symbols.

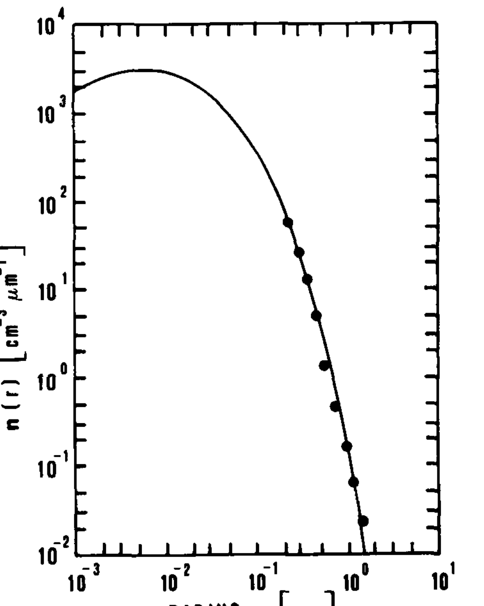

Fig. 2. Example of good-fit solution found on the basis of the modified gamma distribution function for an empirical size spectrum measured by Alkezweeny & Slinn (1972) at Quillayute. The parameter set is given by $\alpha = 1$, $\gamma = 0.3796$, $r_c = 6.714 \times 10^{-3}\,\mu m$ and f_{min} is equal to 0.005.

Alkezweeny & Slinn (1972), Zuyev et al. (1973). The results give four Junge-type solutions with values of ν^* between 1.0 and 3.1. The modified gamma parameter sets are shown in Fig. 3. About 85% of these solutions give values of f_{min} less than 0.1.

(c) *Desert type.* Eight runs measured by Sverdrup et al. (1975) in the Mojave desert during clean air conditions and incursions of aged aerosol have been examined. Two runs correspond to Junge-type solutions with values of ν^* around 2.1. The modified gamma solutions present values of f_{min} less than 0.02. They give an α equal to 1 or 2, while those relative to clean air conditions present γ around 0.2 and r_c of about $7 \times 10^{-4}\,\mu m$ and those affected by incursions of aged aerosol present values of γ around 0.3 and r_c around $4 \times 10^{-3}\,\mu m$.

(d) *Urban and anthropogenic type.* With regard to different urban areas, 107 runs have been taken from: Junge (1955), Cartwright et al. (1956), Kuroiwa (1956), Pasceri & Friedlander (1965), Clark & Whitby (1967), Noll (1967), Pueschel & Noll (1967), Harris (1971), Whitby (1971), Alkezweeny & Slinn (1972), Andreyev et al. (1972), Willeke et al. (1974), Tomasi et al. (1975). In all 26 runs present Junge-type solutions with ν^* ranging from 1.2 to 3.3. The Deirmendjian's function parameter sets with r_c less than 1 μm are given in Fig. 4. The results concerning giant particle modes (Andreyev et al., 1973) are not shown in the figure as they give too high values of γ (from 1.2 to 6.3) and of r_c (from 1.4 to 4.3 μm) with α ranging between 4 and 6. About 44% of the solutions present values of f_{min} less than 0.1, whereas 36% correspond to f_{min} between 0.1 and 0.2.

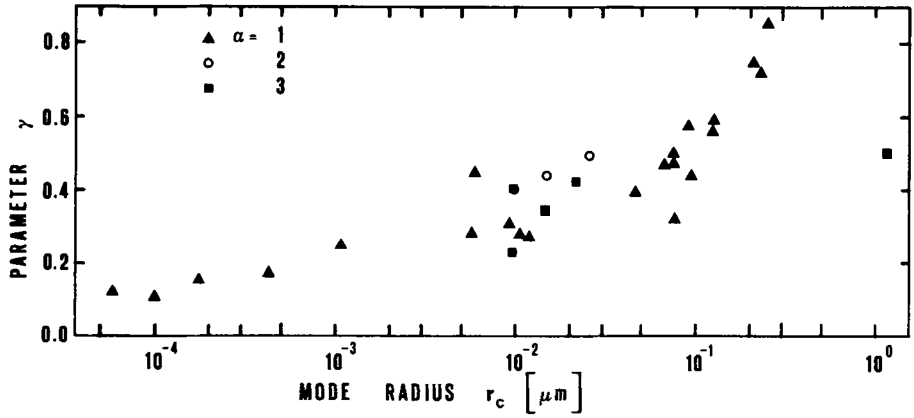

Fig. 3. Sets of the modified gamma size distribution parameters relative to empirical size spectra of continental type particles. The mode radius r_c is given in abscissa, the parameter γ in ordinate while the parameter α is indicated by means of different symbols.

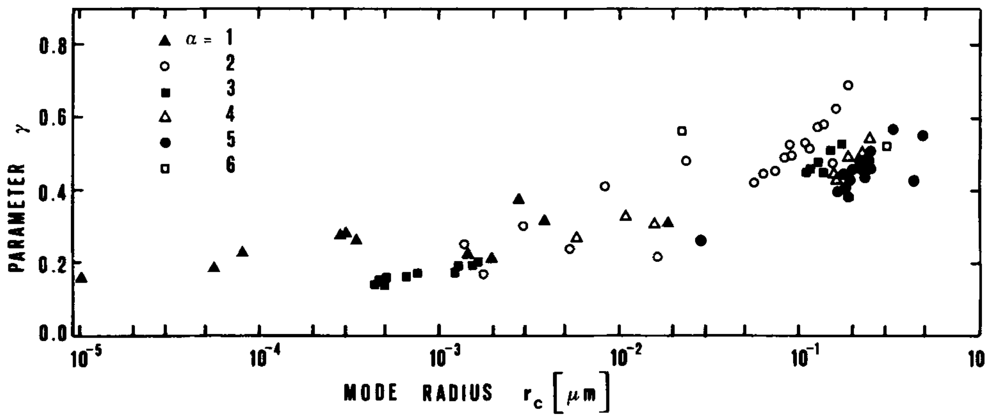

Fig. 4. Sets of the modified gamma size distribution parameters relative to empirical size spectra of urban and anthropogenic type particles. The mode radius r_c is given in abscissa, the parameter γ in ordinate while the parameter α is indicated by means of different symbols.

4. Relationships between skewness and mode radius

The present results give evidence for the wide shape variability which characterizes the atmospheric particle size distributions. Moreover the atmospheric particle load can be generally represented with multimodal features given by modes centred at distinct values of the radius, as shown by Fig. 5 where the empirical size spectrum measured by Zuyev et al. (1973) in the upper troposphere is fitted by four modes based on Deirmendjian's function.

Therefore it appears useful to define a procedure which permits one to immediately find size distribution models centred at any radius of the real range and with shapes which have to be consistent with the features most frequently found in the real atmosphere. For this purpose a significant parameter of the size distribution appears to be the moment coefficient of skewness a_3 which gives a measure of the degree of asymmetry of the distribution curve. Since the coagulation and sedimentation processes are mainly responsible for variations of the airborne particle size distribution with respect to the original shape, the skewness a_3 appears to be a significant parameter related to the efficiency of these mechanisms. With regard to this aspect,

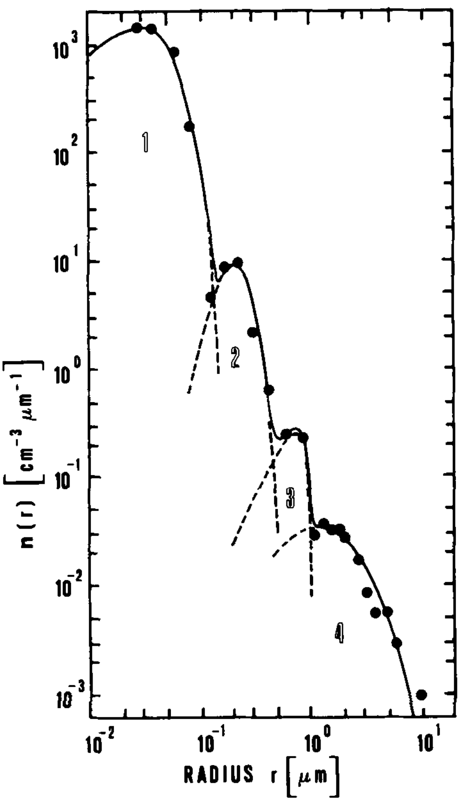

Fig. 5. Example of four-modal best-fit solution given by the computational procedure applied to the particle size spectrum measured by Zuyev et al. (1973) in the upper troposphere.

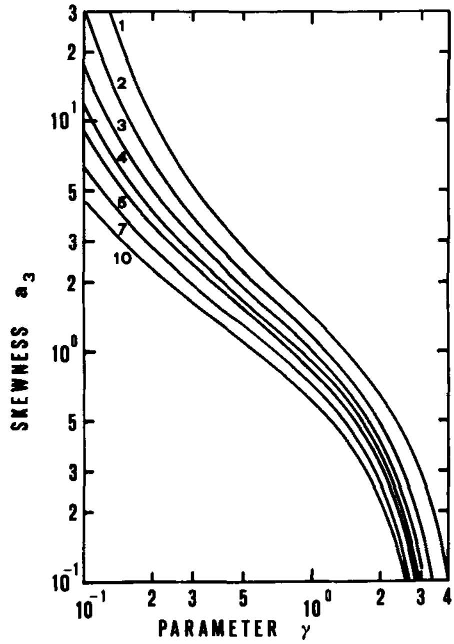

Fig. 6. Moment coefficient of skewness a_3 of the modified gamma distribution function versus the parameter γ for different integer values of the parameter α. For each isopleth the corresponding value of α is given.

Brock (1972) points out that condensation-removal growth processes produce characteristic particle size distributions which are skewed and long-tailed distribution curves.

Related to the modified gamma distribution function the parameter a_3 can be expressed as a function of α and γ. Its expression in terms of the gamma function is given in a previous paper (Tampieri & Tomasi, 1976). Drawn on the basis of this expression the diagram of Fig. 6 permits one to derive the value of a_3 from α and γ, independently on the mode radius r_c.

Taking into account the fact that Figs. 1, 3 and 4 show that the parameters α and γ tend to gradually increase with the mode radius and that the diagram of Fig. 6 gives values of skewness which decrease as α and γ assume higher values, it may be deduced that the skewness should present the general tendency to decrease with the mode radius. Thus the values of skewness a_3 corresponding to the obtained parameter sets have been plotted as a function of the logarithm of the mode radius r_c for the various types of atmospheric particles. For each type, equations describing the trends of the skewness have been found:

(a) *Coastal and maritime type.* The data relative to this type are shown in Fig. 7. The corresponding correlation line is given by

$$a_3 = -0.55 - 1.63 \ln rc \qquad (2)$$

with the correlation coefficient $R = -0.880$.

The most frequent value of α found for this type of particles is 1 (see Fig. 1). Then the following procedure can be adopted for deriving realistic and representative size distribution models based on Deirmendjian's function: eq. (2) gives the possibility of evaluating a_3 for any chosen value of the mode radius. For each value of a_3, the isopleth of the diagram of Fig. 6 which corresponds to the most appropriate value of the parameter α permits one to identify that value of γ which most properly completes the representative parameter set of the modified gamma size distribution.

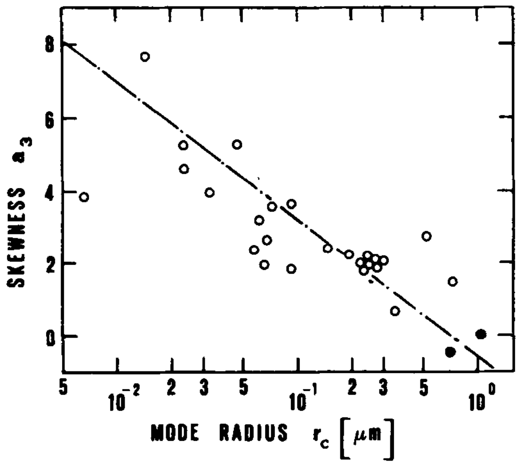

Fig. 7. Moment coefficient of skewness a_3 plotted as a function of the mode radius r_c relative to the solutions found for empirical runs of coastal and maritime type. The correlation line is given by eq. (2). Solid circles refer to the two modes given by Kuroiwa (1956) for sea salt nuclei.

Similarly such a procedure can be applied to the following relationships between skewness and mode radius which are connected with the other types of atmospheric particles.

(b) *Continental type.* The data relative to the parameter sets obtained for continental particles are shown in Fig. 8, distinguishing them into two classes. The first class C.3 includes the data corresponding to samples taken at altitudes higher than 3 km and therefore related to size distribution features proper to the "background" particle population. The corresponding correlation line is defined by

$$a_3 = -0.31 - 1.39 \ln r_c \quad (3)$$

with $R = -0.896$. As can be seen in Fig. 3, a value of α equal to 1 has to be associated with eq. (3).

The second class C.4 refers to samples taken at levels lower than 3 km and therefore probably influenced by local particles sources. The correlation line, given by

$$a_3 = -4.34 - 1.73 \ln r_c \quad (4)$$

with $R = -0.803$, suggests representative size distribution models typical of the atmospheric layer near the ground in unpolluted areas. Fig. 3 suggests the use of $\alpha = 1$ for r_c less than 0.1 μm and of $\alpha = 3$ for the larger mode radii.

(c) *Desert type.* The data relative to the empirical size spectra measured in the Mojave desert are also plotted in Fig. 8. The correlation line gives

$$a_3 = -4.52 - 1.64 \ln r_c \quad (5)$$

with $R = -0.950$. Taking into account that a large part of data are affected by incursions of aged aerosol the similarity of trend of eq. (5) and eq. (4) is not surprising.

(d) *Urban and anthropogenic type.* The data of this type have been plotted in Fig. 9 which gives evidence for separating them into three classes.

The class U.6 includes size distributions in which the high values of the skewness may be due to the

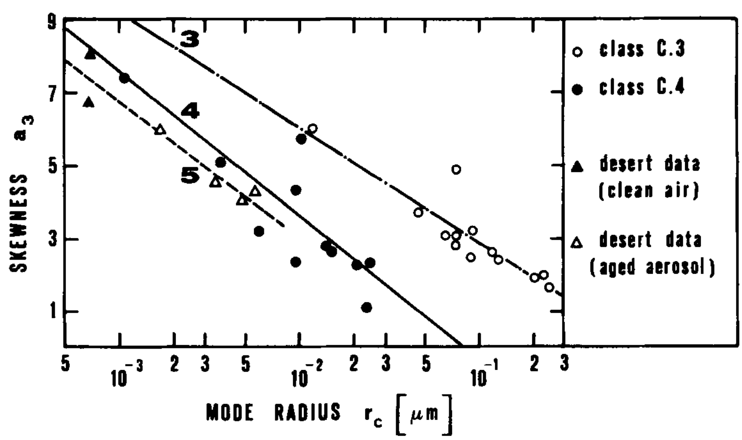

Fig. 8. Moment coefficient of skewness a_3 plotted as a function of the mode radius r_c relative to the solutions found for empirical runs of continental and desert type. The correlation lines 3, 4 and 5 are given respectively by eqs. (3), (4) and (5).

continuous production of very small particles. Its correlation line is given by

$$a_3 = -1.14 - 1.54\, ln\, r_c \qquad (6)$$

with $R = -0.980$. From the examination of the corresponding parameter sets, appropriate values for α result to be 1 and 2.

The class U.7 includes the size distributions characterized by relatively lower values of a_3 and typical of the great urban centres. The correlation line is given by

$$a_3 = -2.18 - 1.12 \ln r_c \qquad (7)$$

with $R = -0.907$. The value of α equal to 3 can be associated with this relationship.

The class U.8 refers to size distributions characterized by predominant contents of large and giant particles. Its data present a skewness which varies slightly with respect to the mode radius and assure almost symmetrical shapes in the resulting modes. The correlation line is described by

$$a_3 = 0.80 - 0.58 \ln r_c \qquad (8)$$

with $R = -0.961$. Correspondingly, Fig. 4 suggests that the parameter α has to assume values from 2 to 5 as the mode radius gradually increases. As can be seen in Fig. 9, the solution for the combustion product data proposed by Kuroiwa (1956) and that derived from the data of Noll (1967) for haze conditions perfectly agree with the skewness trend described by eq. (8).

5. Conclusions

The continuity which characterizes the evolutionary trend of the atmospheric particle load and the wide variability generally displayed by the particle size distributions of the real atmosphere call for defining representative models covering the whole range of the mode radius. For this purpose the proposed relationships between the skewness and the mode radius supplied by the correspondingly suggested values of the parameter α represent a useful tool for obtaining various and representative models in terms of Deirmendjian's function for each atmosphere type. These relationships are characterized by different angular coefficients and give evidence for the different variability of the degree of asymmetry with respect to the mode radius. Generally each single equation gives size distributions with the smaller particle wing which drops more rapidly as the mode radius increases while the shapes of the right wing present more limited variations. Such a behaviour appears to be consistent with the variable properties of the growth processes which affect the small particles

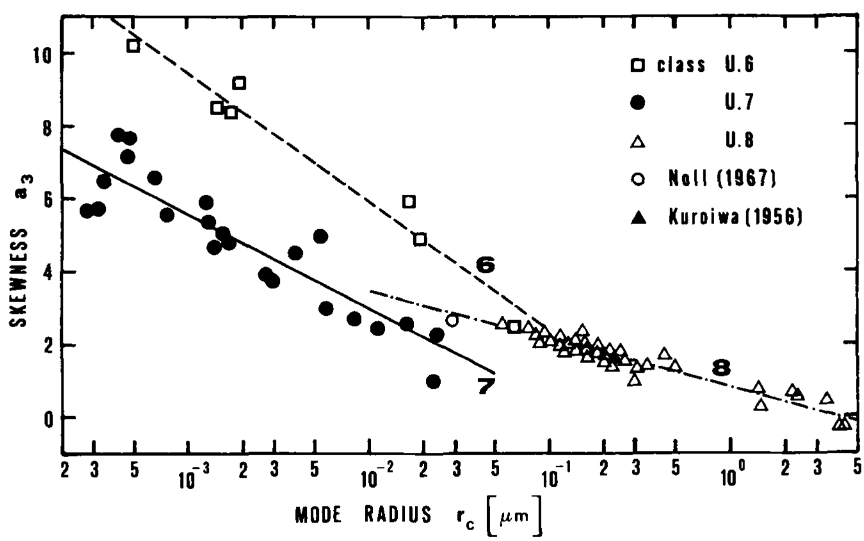

Fig. 9. Moment coefficient of skewness a_3 plotted as a function of the mode radius r_c relative to the solutions found for empirical runs of urban and anthropogenic type. The correlation lines 6, 7 and 8 are given respectively by eqs. (6), (7) and (8).

and agrees well with a rather marked efficiency both of the growth by condensation and of sedimentation and the other removal mechanisms in shaping the large-particle wing of the size distribution.

By following the present procedure, sets of representative size distribution models centred at well-spaced values of the radius can be derived for atmospheric particles of different origin. These models can be applied to the electromagnetic theory for evaluating the scattering and absorption of radiation by various atmospheric particle loads.

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РАСПРЕДЕЛЕНИЯ ПО РАЗМЕРАМ ТРОПОСФЕРИЧЕСКИХ ЧАСТИЦ В ТЕРМИНАХ МОДИФИЦИРОВАННОЙ ФУНКЦИИ ГАММ И В СООТНОШЕНИЯХ МЕЖДУ АССИМЕТРИЕЙ И ДАЛЬНОГО РАДИУСА

С помощью численных методов в терминах модифицированной функции гамма-распределения интерпретируются спектры размеров тропосферных частиц, находящихся в воздухе. Получены результаты относящиеся к частицам разлиуного происхождения: берегового и морского, континентального, пустынного, городского и антропогенного. Обнаружены корреляции между асимметрией полученных распределений размеров и логарифмом радиуса соответствующей моды для различных типов атмосферных частиц. На основе полученных соотношений предлагается метод, пригодный для получения распределения размеров частиц вещества, типичных для различных атмосфер.