

GEOPHYSICAL BULLETIN

No. 21

**On the Particle Size Analysis
of Polydisperse Aerosols using a Diffusion Battery
and the Exhaustion Method**

by

A. L. METNIEKS and L. W. POLLAK

Scoil na Fisice Cosmaí, Institiúid Árd-Léinn Bhaile Átha Cliath
School of Cosmic Physics, Dublin Institute for Advanced Studies

April 1962

Price : 7s. 6d.

CONTENTS

ABSTRACT

INTRODUCTION

THE DYNAMIC METHOD

Homogeneous Aerosols
Heterogeneous Aerosols

RESOLUTION OF HETEROGENEOUS AEROSOLS, EXHAUSTION METHOD

Procedure

Numerical Examples

- (i) Heterogeneous aerosol containing three components
- (ii) Heterogeneous aerosol containing nine components with Gaussian distribution of their concentration

Actual Example

- (i) Equipment
- (ii) Artificially produced aerosol stored in large gasometer

RESULTS OF 91 RESOLUTIONS OF POLYDISPERSE AEROSOLS produced and stored in large gasometer

APPENDICES

I A : Computation of 'observed' (apparent) diffusion coefficient as a function of air-flow. - Mixture of three aerosols

I B' : Resolution of heterogeneous aerosol using the Exhaustion Method. - Mixture of three aerosols

I B" : Resolution of heterogeneous aerosol using the Exhaustion Method (Concluded). - Mixture of three aerosols

II A : Computation of 'observed' (apparent) diffusion coefficient as a function of air-flow. - Mixture of nine aerosols with Gaussian distribution of their concentration

II B' : Resolution of heterogeneous aerosol using the Exhaustion Method. - Mixture of nine aerosols with Gaussian distribution of their concentration

II B" : Resolution of heterogeneous aerosol using the Exhaustion Method (Concluded). - Mixture of nine aerosols with Gaussian distribution of their concentration

III A : Measurements for resolution of heterogeneous aerosol using the Exhaustion Method on 21 January 1962, interpolated for 11h 53m. - Artificially produced aerosol stored in large gasometer.

III B': Resolution of heterogeneous aerosol using the Exhaustion Method. - Artificially produced aerosol stored in large gasometer; Measurements on 21 January 1962, interpolated for 11h 53m

III B": Resolution of heterogeneous aerosol using the Exhaustion Method (Concluded). - Artificially produced aerosol stored in large gasometer; Measurements on 21 January 1962, interpolated for 11h 53m

FIGURES

1 : Heterogeneous aerosol containing nine components with Gaussian distribution of their concentration

2a: Point clouds of first components of young and old aerosols produced and stored in large gasometer

2b: Point clouds of second components of young and old aerosols produced and stored in large gasometer

2c: Point clouds of third components of young and old aerosols produced and stored in large gasometer

ABSTRACT

The results of 91 resolutions of polydisperse aerosols produced and stored in a large gasometer using the Exhaustion Method are given and discussed.

Details are given of the equipment required and of the method employed. The interpretation of the results obtained is illustrated by numerical and actual examples.

To facilitate the application of the Exhaustion Method all necessary computations for the size analysis of three examples are compiled in the Appendices.

INTRODUCTION

In the course of various experimental investigations (^{1,2,3,4}) in which the average radius of the condensation nuclei of a stored aerosol was required, we have carried out numerous particle size analyses of polydisperse aerosols using a diffusion battery and the 'Exhaustion Method'(⁵). Since these analyses enable one to study the dependence of the heterogeneity of a stored aerosol on the mode of production of the nuclei, their age and concentration, we are publishing in the following the results of 91 resolutions together with a detailed discussion of the method employed.

THE DYNAMIC METHOD

Homogeneous Aerosols - This method of determining the diffusion coefficient of condensation nuclei was devised by the late J. J. NOLAN and V. H. GUERRINI (⁹). The first correct solution of the underlying diffusion problem was given by C. H. BOSANQUET. His formulae were afterwards confirmed by P. G. GORMLEY (¹⁰) and re-derived by W. De MARCUS & J. W. THOMAS (¹¹) in 1952. The coefficients in the formulae of all the authors mentioned agree very closely and can be accepted as correct.

In the dynamic method the nuclei are passed through a 'box' containing a number of vertical rectangular channels arranged in parallel. This diffusion apparatus originally called diffusion box is now known under the name diffusion battery(¹¹). The theory as developed by the authors mentioned above, assumes that the air-flow is parallel to the length of the channel and that

there is only one diffusion coefficient of the nuclei or, in other words, that the aerosol is homogeneous with respect to particle size. Further assumptions are: All particles which contact the wall of the channel adhere, the effect of turbulence in the entrance to the channels is negligible, the pressure drop across the battery is small and the diffusion of the particles is not affected by electrostatic charges on the particles or battery wall⁽¹⁾.

The nucleus concentration Z at the entrance of the channel and the concentration Z_v at its exit are measured. The ratio of these concentrations Z_v/Z is related to the diffusion coefficient D of the nuclei, the volume of the aerosol (air containing the nuclei) q passing per second through each channel and the dimensions of the channel (L = length, b = height and $2a$ = width) by the following formula

$$Z_v/Z = 0.9099 \exp(-x) + 0.0531 \exp(-11.369x) \quad \dots (1)$$

where

$$x = 3.77 b L D a^{-1} q^{-1} \quad \dots (2)$$

If c is the number of channels in the diffusion apparatus, the volume of the aerosol passing per second through the diffusion battery $Q = cq$ and

$$x = 3.77 b c L D a^{-1} Q^{-1} \quad \dots (3)$$

or

$$x = D/fQ \quad \dots (4)$$

where

$$f = 3.77^{-1} b^{-1} c^{-1} L^{-1} a \quad \dots (5)$$

If we assume x of such value that the second term in Equ.(1) can be ne-

glected

$$Z_v/Z = 0.9099 \exp(-x)$$

... (11)

Up to 1955 the air-flow Q through the diffusion box was usually adjusted in practice so that the ratio Z_v/Z had a value of between 0.7 and 0.2 and the duration of a complete experiment was not unduly long (14).

It has been shown (6) that within the range of 1 to 4 litres/min air-flow a change by 1 litre/min alters the diffusion coefficient by roughly 12 %. Since for the technical reasons mentioned above during one experiment adjustments of the air-flow by two litres/min were not uncommon up to 1956, the diffusion coefficient may be wrong by 25% of its value for this reason alone.

Heterogeneous Aerosols - In 1955 P O L L A K and his co-workers discovered a marked dependence of the apparent diffusion coefficient D on the air-flow Q through the diffusion apparatus (6,7) as determined with the diffusion battery, using G O R M L E Y's formula. They found a regular and systematic increase of D with increase of Q .

This apparent increase of the diffusion coefficient with increasing flow rates had been found previously and the observation published in 1955 (12,13) but the publication came to our notice only in 1957.

M E T N I E K S deduced from G O R M L E Y's formula that such an effect must appear when the aerosol is heterogeneous with respect to particle size. P O L L A K in turn recognised the potentialities of this explanation and suggested the 'Exhaustion Method' (5) which is similar to the determination of hidden periodicities (15).

The influence of heterogeneity on the determination of the diffusion coefficient by the dynamic method was investigated numerically for polydisperse

aerosols consisting of a mixture of components without ⁽⁵⁾ and with various Gaussian particle size distribution ⁽⁸⁾.

The theory of the dynamic method for heterogeneous aerosols as developed quite generally in Ref. 5 gave the following results.

(i) The diffusion coefficient D of a mixture of nuclei depends on the air-flow Q and also on the dimensions of the diffusion apparatus used.

(ii) If we measure by the dynamic method, for different air-flows, the diffusion coefficient D of an aerosol containing a mixture of condensation nuclei of various sizes, then the graph representing D as a function of Q intersects with the ordinate (D) axis at a value D_s which gives the smallest diffusion coefficient in the mixture.

(iii) With increasing air-flow the apparent diffusion coefficient approaches the weighted average of the component diffusion coefficients, the weights being their partial concentrations.

(iv) The apparent D as measured by the dynamic method is always smaller than the arithmetic mean of the diffusion coefficients of the components and only for infinite air-flow does it approach the weighted mean.

(v) The apparent diffusion coefficient of a mixture of nuclei as determined by the dynamic method, particularly when low air-flow is used, has little meaning. The same apparent diffusion coefficient can be found with the same air-flow for mixtures of nuclei having completely different partial concentrations and sizes of their components.

RESOLUTION OF HETEROGENEOUS AEROSOLS

EXHAUSTION METHOD

Procedure - This method assumes a polydisperse aerosol with an unknown

number of components and makes no assumption regarding their number. The respective (partial) concentrations of the components are $Z^{(i)}$, their proportions $p^{(i)}$ and diffusion coefficients $D^{(i)}$.

We have measured the apparent diffusion coefficient for various and particularly for low air-flows. Then, as mentioned above and proved in Ref.5, the measured D converge towards D^* i.e. the smallest diffusion coefficient in the composite aerosol, when Q approaches zero. The intersection of the curve representing the measured values of D as a function of the air-flow Q with the ordinate (D) axis or a calculation of the ordinate in the origin gives D^* .

For determining the p^* of this aerosol component with the lowest diffusion coefficient (or the largest nuclei) we use Equ.15 deduced in Ref.5

$$p^* = \exp(-D/fQ) : \exp(-D^*/fQ) \quad \dots (6)$$

Equ.6 is valid only for sufficiently small values of Q and on the assumption that none of the $p^{(i)}$ is infinitesimal.

Now, we remove this component (D^* , p^*) from the measured Z_v/Z values, treating the remainder as the new Z_v/Z from which we compute a set of new values of D which we call D^* . These new diffusion coefficients D^* are plotted as a function of Q , from the intersection of which with the ordinate axis we obtain D'' .

In order to compute the D^* values, we form

$$\begin{aligned} (Z_v - Z_v^*) / (Z - Z^*) &= 0.9099 \left[\exp(-D/fQ) - p^* \exp(-D^*/fQ) \right] : (1 - p^*) \\ &= 0.9099 \exp(-D^*/fQ) \end{aligned} \quad \dots (7)$$

or, in practice, we use tables for Z_v/Z vs x which take also the second term into consideration.

With Equ.(6) which now reads

$$p'' = (1 - p^*) \exp(-D^*/fQ) : \exp(-D''/fQ) \dots (8)$$

we get p''' and so on.

Numerical Examples - (i) Heterogeneous aerosol containing three components. In Ref.5 the exhaustion method was applied to a composite aerosol made up of three components with equal concentrations $p^* = p'' = p''' = 0.333$ and with the corresponding diffusion coefficients $D^* = 20$, $D'' = 100$, $D''' = 200$. *) The constant f of the diffusion apparatus was assumed to be 46.25 (Diffusion Battery No.1 of the School of Cosmic Physics).

The resolution gives

$$D^* = 20, \quad p^* = 0.333 \text{ (from Equ.6 for } Q = 0.2\text{)} ;$$

$$D'' = 99, \quad p'' = 0.341 \text{ (from Equ.8 for } Q = 0.5\text{)} ;$$

$$D''' = 207, \quad p''' = 0.326$$

The calculated diffusion coefficients agree very well indeed with the 'observed', the maximum difference being 4%. The partial concentrations found by the exhaustion method approach the actual proportions of the components within 3%.

*) The diffusion coefficients are given without the factor 10^{-6} (cm^2/sec), the radii without the factor 10^{-6} (cm) and the constant f of the diffusion batteries without the factor 10^{-6} .

All computations required for the construction of the composite aerosol and its resolution are given in Appendix I.

(ii) Heterogeneous aerosol containing nine components with Gaussian distribution of their concentration. We assume a composite aerosol of $Z = 10000$ nuclei/cm³ and nine components as defined in Table 1. The average diffusion coefficient of the aerosol $\bar{D} = 220$, the average radius $\bar{r} = 0.856$.

Two somewhat different analyses gave the following results.

Analysis 1 :

$$\begin{array}{lll} D' = 62 & D'' = 155 & D''' = 301 \\ p' = 0.020 & p'' = 0.503 & p''' = 0.477 \end{array}$$

$$\bar{D} = D'p' + D''p'' + D'''p''' = 222$$

$$\bar{r} = r'p' + r''p'' + r'''p''' = 0.860$$

Analysis 2 :

$$\begin{array}{lll} D' = 62 & D'' = 140 & D''' = 259 \\ p' = 0.020 & p'' = 0.302 & p''' = 0.678 \end{array}$$

$$\bar{D} = 219$$

$$\bar{r} = 0.857$$

We see that using the Exhaustion Method for the analysis of an aerosol containing nine components with approximately Gaussian distribution of their concentration, we obtain only three components, even when the D -values are accurately known. Thus, the Exhaustion Method is a numerical spectrometer of limited resolution power. We must, therefore, expect that an actual aerosol can in general be resolved by this method into three components only.

Table 1 - Heterogeneous aerosol containing nine components

with Gaussian distribution of their concentration; $Z = 10000 \text{ nuclei/cm}^3$.

i	1	2	3	4	5	6	7	8	9
$D_i \cdot 10^6$	60	100	140	180	220	260	300	340	380
Z_i	140	475	1170	2010	2410	2010	1170	475	140
$P_i = Z_i / Z (\%)$	1.40	4.75	11.70	20.10	24.10	20.10	11.70	4.75	1.40
$M_i \cdot 10^6 \text{ cm}$	2400	4000	5600	7200	8800	10400	12000	13600	15200
$r_i \cdot 10^6 \text{ cm}$	1.605	1.229	1.034	0.9087	0.8200	0.7549	0.7020	0.6588	0.6210
$\frac{1}{100} P_i r_i$	0.0225	0.0584	0.1210	0.1826	0.1976	0.1517	0.0821	0.0313	0.0087

Notes:

- (i) Mobility $M_i = 40 D_i$
- (ii) r_i from tables Radius vs. Mobility in Geophys. Bull. No.19 of the School of Cosmic Physics, Dublin (Ref.17) for 20°C
- (iii) The averages of D and r are

$$\bar{D} = \frac{1}{100} \sum P_i D_i = 220$$

$$\bar{r} = \frac{1}{100} \sum P_i r_i = 0.856$$

Since these three 'bands' replace the nine original components, they lump them together. From these few bands, found by the Exhaustion Method, we compute, however, the same average diffusion coefficient \bar{D} and the same average radius \bar{r} as from the original components. The numerical example above shows that the agreement between the \bar{D} and \bar{r} computed from the D and r

found by analysis and calculated from the D and r of the original aerosol is better than 1%.

Fig.1 illustrates the situation. It shows the histogram of the frequencies Z_i vs the diffusion coefficient $D_i \cdot 10^6$ of Table 1, the ogive (cumulative frequencies) of the Z_i , the ogive of the three components found by the Exhaustion Method and the histogram of these components. This histogram which is determined by the histogram of the original Z_i and their ogive, shows how the original frequencies are lumped together by the components obtained by the analysis.

In Appendix II all computations required for the construction of the composite aerosol and its resolution are given.

Actual Example - (i) Equipment required: A photo-electric nucleus counter^(16,18) without or with compensating circuit for measuring very low concentrations of condensation nuclei.

A diffusion battery preferably one without end-pieces or connecting tubing⁽²⁰⁾. The elimination of the end-pieces and connecting tubing removes any end-effect and diffusion losses in these parts of the diffusion battery and reduces the volume of the diffusion apparatus to that of its channels, thus minimising the lag in the response of the battery. Since the volume of the fog-tube of a photo-electric counter with 2.5 cm air-column diameter is only 288 cm³, a battery without end-pieces shortens the filling operation so much that air-flows down to 0.2 litres/min are possible without unduly lengthening the measurement. These low air-flows are particularly valuable in the application of the exhaustion method for the size resolution of polydisperse aerosols.

The diffusion batteries used in this investigation were: Nos. I & III and

BWE 1 (G.E., Schenectady). Particulars of the batteries together with the range of D for various Q's for Z_v/Z within the limits 0°675 and 0°25 are given in Table II. In the Exhaustion Method values of Z_v/Z as small as 0°01 proved useful and indispensable.

(ii) Artificially produced aerosol stored in large gasometer - The nuclei used in this experiment were produced at 9h 35m by heating a nichrome wire helix mounted on a pyrex glass tube in the centre of a Mylar balloon of 4200 litres content with 4°20 A for 60 sec.

In Table III (a) the actual measurements are compiled and in Table III (b) are given the Z & Z_v readings interpolated for 11h 53m from graphs of the original measurements (Table III a). These values form the starting point of the resolution and are transferred to Cols. 1 to 3 in Appendix III A.

Appendix III shows all details of the resolution which gives

$$\begin{array}{ll} D' = 26, & p' = 0°050 \\ D'' = 300, & p'' = 0°592 \\ D''' = 840, & p''' = 0°358 \end{array}$$

and average radius of this aerosol $\bar{r} = 0°690 \cdot 10^{-6}$ cm.

RESULTS OF 91 RESOLUTIONS OF POLYDISPERSE AEROSOLS

produced and stored in a large gasometer

In Table IV are given, arranged according to increasing average radius, the results of our resolutions of polydisperse aerosols, produced and stored in a large rubber or metallised Mylar balloon gasometer, up to 20th February 1962.

Table II - Particulars of diffusion batteries.

Diff. Battery	No. 1				No. 3				BWE 1 (G.E., Schenectady)		
Plates	Glass				Glass				Metal		
Separators	Cardboard				Cardboard				Metal		
L cm	50.2				40.04				50.0		
b cm	8.0				19.07				7.972		
2a cm	0.084				0.0466				0.0529		
c	10				30				5		
Constant	$2.77405 \cdot 10^{-6}$				$2.69804 \cdot 10^{-7}$				$3.52028 \cdot 10^{-6}$		
Volume (cm^3)											
(i) Channels	337				1067				80		
(ii) End-pieces	318				2515				0		
(iii) Total	655				3582				80		
Q (litres/min)	$D \cdot 10^6$			$fQ \cdot 10^6$	$D \cdot 10^6$			$fQ \cdot 10^6$	$D \cdot 10^6$		
	Z_v/Z =0.675	Z_v/Z =0.250	Z_v/Z =0.675		Z_v/Z =0.250	Z_v/Z =0.675	Z_v/Z =0.250				
	$x = 0.3$	$x = 1.3$	$x = 0.3$		$x = 1.3$	$x = 0.3$	$x = 1.3$				
0.5	6.9	30.1	23.117		0.7	2.9	2.248		8.8	38.1	29.335
1.0	13.9	60.1	46.234		1.3	5.8	4.497		17.6	76.3	58.67
2.0	27.7	120.2	92.468		2.7	11.7	8.993		35.2	152.5	117.34
3.0	41.6	180.3	138.702		4.0	17.5	13.490		52.8	228.8	176.01
4.0	55.5	240.4	184.937		5.4	23.4	17.987		70.4	305.1	234.68
5.0	69.4	300.5	231.171		6.7	29.2	22.484		88.0	381.4	293.35
7.0	97.1	420.7	323.639		9.4	40.9	31.477		123.2	533.9	410.69
10.0	138.7	601.0	462.342		13.5	58.5	44.967		176.0	762.7	586.70
14.0	194.2	841.5	647.278		18.9	81.8	62.954		246.4	1067.8	821.38
20.0	277.4	1202.0	924.684		27.0	116.9	89.935		352.0	1525.4	1173.40

Notes:

(i) $D = x \cdot \text{Const. } 1000/60$ (for $Q = 1$ litre/min)

(ii) $D = xfQ$

Table III - Measurements with diffusion battery BWE 1 (G.E., Schenectady)
on 21 January 1962.

(a)

Time	Q	Counter of BWE 1 Reading		Time	Q	Counter of BWE 1 Reading	
		Z	Z_V			Z	Z_V
h m	litres/min	%	%	h m	litres/min	%	%
11 36	20	46°3		12 21	20	54°1	
38	20	46°9		23	20		60°6
40	20		54°2	25	10		65°5
42	10		59°4	27	5		72°7
44	5		68°1	29	20	56°0	
46	20	48°3		32	2°5		81°7
49	2°5		78°1	36	1		91°2
53	1		89°5	42	0°5		93°1
59	0°5		92°4	44	20	58°0	
12 01	20	51°0					

(b)

Q	Counter of BWE 1 Interpol. Reading for 11h 53m	
	Z	Z_V
litres/min	49°5	
0°5		92°3
1		89°5
2°5		78°5
5		69°1
10		61°1
20		56°2

Up to the 1st December 1961 the nuclei used were produced by heating a length of nichrome resistance wire (a helix of a few turns) suspended in the axis of a Tufnol 'boat' which is mounted approx. in the centre of the gasometer. The nuclei, therefore, contain an admixture of burned Tufnol.

After the 1st December 1961 nuclei were produced by heating an identical helix of nichrome wire mounted on a pyrex glass tube. Nuclei of high concentration generated in this way require very much longer heating duration and their size is small.

In all experiments with serial numbers 6 and more the nuclei after production were diluted with filtered air to ensure good mixing in the balloon gasometer and to slow down decay. In the experiments with serial numbers 1 to 5 the dilution with filtered air took place during the production of nuclei in order to obtain very small nuclei.

In the Column headed 'Time' (Production of Nuclei) the hours shown refer to the corresponding day given in Column 'Date' when positive, to the previous day when negative; e.g. for the experiment on 20th October 1959 (Serial Number 74) the nuclei used were produced on the 19th at 1700, therefore in the Column 'Time' of the production of nuclei is entered -7 hrs.

With regard to the accuracy of the resolution results it should be noted that there is a certain latitude in the extrapolation to zero air-flow in determining the D^* and D'' and in the interpretation of the graphs. Two examples selected at random and reproduced below (page 24), are intended to show that in spite of this, two independent analysts can arrive at essentially the same components.

The agreement of the average radius \bar{r} is particularly good.

Table IV - Resolutions of polydisperse aerosols

Serial No.	Date	Production of Nuclei			Nuclei		Components of Aerosol			
		Time	Current	Duration	Age	Concentration	Diffusion Coefficient			
							D ^I	D ^{II}	D ^{III}	D ^{IV}
		h	A	sec	h	Z	D ⁽ⁱ⁾ . 10 ⁶			
1	24- 4-61	10°0	4°90	30	4°6	973	500	1800		
2	27- 4-61	10°0	4°80	150	4°6	2854	1200			
3	28- 4-61	9°5	4°80	900	5°6	3160	200	620		
4	28- 4-61	9°5	4°80	900	6°3	2592	210	620		
5	28- 4-61	9°5	4°80	900	6°9	2147	210	620		
6	13- 2-62	9°6	4°25	120	1°9	11682	25	335	895	
7	21- 1-62	9°6	4°20	60	2°3	11492	26	300	840	
8	6- 2-61	14°3	4°70	12	1°9	24871	180	550		
9	13- 2-62	9°6	4°25	120	2°6	6912	29	320	785	
10	4- 9-61	9°5	4°85	25	2°4	10972	30	220	400	800
11	21- 1-62	9°6	4°20	60	3°0	6984	20	300	720	
12	10- 2-61	10°2	4°75	12	2°3	13370	100	270	600	
13	1- 3-61	9°6	4°90	15	1°7	20811	35	150	300	606
14	18- 8-61	9°7	4°90	25	2°3	27295	80	250	790	
15	20- 2-62	9°9	4°25	180	2°1	18852	58	279	900	
16	21- 8-61	9°9	4°90	25	2°1	32330	60	190	500	
17	2- 7-59	9°7	4°95	5	2°3	7700	80	280	560	
18	16- 2-62	9°5	4°25	180	2°5	12154	69	256	740	
19	6- 3-61	9°6	4°90	18	1°7	23124	50	180	334	
20	30- 8-61	10°6	4°80	35	1°9	31489	60	170	350	
21	3- 2-62	9°5	4°30	240	2°0	17764	74	259	850	
22	20- 2-62	9°9	4°25	180	2°9	11118	52	260	950	
23	29- 8-61	9°6	4°85	30	2°9	21203	40	150	250	825
24	16- 1-62	10°0	4°20	30	2°0	13547	15	290	910	
25	1- 2-61	14°2	4°50	10	1°8	22972	80	400		

produced and stored in large gasometer.

Components of Aerosol				Average		Diff. Coeff. to Average Radius	Min. Z_v/Z	Diffusion Battery	Serial No.				
Proportion				Diffusion Coefficient	Radius								
p ^I	p ^{II}	p ^{III}	p ^{IV}										
				$\bar{D} \cdot 10^6$	$\bar{r} \cdot 10^6$	$D_{\bar{r}} \cdot 10^6$							
0°060	0°940			1722	0°30	1620	0°004	BWE	1				
1°000				1200	0°35	1200	0°005	BWE	2				
0°190	0°810			540	0°56	475	0°006	BWE	3				
0°280	0°720			505	0°58	430	0°007	BWE	4				
0°340	0°660			480	0°61	400	0°009	BWE	5				
0°035	0°640	0°325		506	0°65	354	0°014	BWE	6				
0°050	0°592	0°358		480	0°69	312	0°019	BWE	7				
0°470	0°530			376	0°70	300	0°020	BWE	8				
0°057	0°680	0°263		426	0°71	295	0°020	BWE	9				
0°025	0°431	0°440	0°104	355	0°72	282	0°008	BWE	10				
0°057	0°653	0°290		406	0°75	262	0°026	BWE	11				
0°140	0°640	0°220		319	0°76	257	0°004	BWE	12				
0°020	0°225	0°607	0°148	306	0°77	250	0°001	BWE	13				
0°151	0°683	0°166		314	0°81	225	0°009	BWE	14				
0°135	0°731	0°134		333	0°81	227	0°017	BWE	15				
0°068	0°611	0°321		281	0°82	220	0°008	BWE	16				
0°305	0°240	0°455		346	0°83	215	0°049	I	17				
0°182	0°723	0°095		268	0°86	200	0°016	BWE	18				
0°065	0°600	0°335		223	0°88	190	0°001	BWE	19				
0°093	0°488	0°419		235	0°88	191	0°011	BWE	20				
0°230	0°674	0°096		274	0°88	191	0°017	BWE	21				
0°166	0°739	0°095		291	0°88	191	0°026	BWE	22				
0°085	0°444	0°277	0°194	299	0°89	187	0°020	BWE	23				
0°108	0°552	0°340		471	0°89	186	0°059	BWE	24				
0°380	0°620			278	0°90	183	0°088	BWE	25				

Table IV - Resolutions of polydisperse aerosols

Serial No.	Date	Production of Nuclei			Nuclei		Components of Aerosol			
		Time	Current	Duration	Age	Concentration	Diffusion Coefficient			
							D ^I	D ^{II}	D ^{III}	D ^{IV}
26	14- 3-61	9°6	4°90	21	1°7	26227	37	190	340	
27	19- 5-61	11°5	4°80	50	1°3	52009	70	160	350	
28	16- 2-62	9°5	4°25	180	3°3	8175	70	280		
29	6- 2-62	9°6	4°15	360	1°7	24542	76	240	700	
30	19- 1-62	9°6	4°20	45	2°0	8456	13	560		
31	3- 2-62	9°5	4°30	240	2°8	11192	69	272		
32	11- 8-61	9°7	4°90	30	2°5	23900	67	230	530	
33	17- 8-61	9°7	4°95	30	2°6	25037	60	194	838	
34	23- 8-61	9°5	4°90	30	2°9	21507	50	150	330	
35	16- 3-61	9°7	4°85	30	1°7	30565	45	130	270	
36	16- 1-62	10°0	4°20	30	2°6	10440	15	330	830	
37	6- 2-62	9°6	4°15	360	2°5	14325	76	218	1250	
38	24- 8-61	9°5	4°85	40	2°6	26054	60	150	320	
39	22- 2-61	9°7	4°85	25	1°7	26140	45	130	303	
40	22- 8-61	9°5	5°00	30	2°7	24542	60	180	400	
41	24- 2-61	9°7	4°85	25	2°4	17415	37	117	257	
42	25- 1-62	9°6	4°25	300	2°3	21087	50	200		
43	27- 7-61	11°7	4°85	30	2°7	21936	60	230		
44	22- 3-61	9°6	4°90	30	2°0	21087	40	100	250	
45	31- 8-61	9°6	1°95	25	6°1	3849	50	150		
46	14- 8-61	10°1	4°85	60	2°1	33300	60	190		
47	2- 8-61	10°4	4°90	45	2°1	32012	60	185		
48	25- 1-62	9°6	4°25	300	3°1	14372	45	135	1000	
49	20- 2-61	9°7	4°80	30	0°8	47731	50	170		
50	20- 2-61	9°7	4°80	30	1°4	33082	50	190		

produced and stored in large gasometer (continued).

Components of Aerosol				Average		Diff. Coeff. to Average Radius	Min. Z_v/Z	Diffusion Battery	Serial No.				
Proportion				Diffusion Coefficient	Radius								
p ^t	p ["]	p ^{'''}	p ^{''''}										
				$\bar{D} \cdot 10^6$	$\bar{r} \cdot 10^6$	$D_{\bar{r}} \cdot 10^6$							
0°047	0°780	0°173		209	0°90	183	0°002	BWE	26				
0°166	0°410	0°424		226	0°92	176	0°014	BWE	27				
0°257	0°743			226	0°92	175	0°022	BWE	28				
0°270	0°646	0°080		235	0°93	173	0°019	BWE	29				
0°138	0°862			485	0°94	168	0°082	BWE	30				
0°261	0°739			219	0°94	170	0°023	BWE	31				
0°260	0°625	0°115		222	0°95	165	0°025	BWE	32				
0°159	0°747	0°094		233	0°95	165	0°019	BWE	33				
0°103	0°532	0°365		205	0°96	162	0°017	BWE	34				
0°065	0°470	0°465		190	0°97	159	0°001	BWE	35				
0°148	0°647	0°205		386	1°02	145	0°081	BWE	36				
0°387	0°536	0°077		202	1°02	145	0°027	BWE	37				
0°233	0°475	0°292		179	1°05	137	0°027	BWE	38				
0°130	0°594	0°276		167	1°07	130	0°003	BWE	39				
0°275	0°625	0°100		169	1°07	131	0°032	BWE	40				
0°085	0°550	0°365		161	1°08	130	0°003	BWE	41				
0°267	0°733			160	1°10	124	0°044	BWE	42				
0°397	0°603			163	1°12	119	0°047	BWE	43				
0°110	0°510	0°380		150	1°14	115	0°003	BWE	44				
0°215	0°785			129	1°16	112	0°036	BWE	45				
0°413	0°587			136	1°18	109	0°049	BWE	46				
0°415	0°585			133	1°19	107	0°049	BWE	47				
0°275	0°645	0°080		179	1°22	101	0°054	BWE	48				
0°451	0°549			116	1°31	88°5	0°075	BWE	49				
0°496	0°504			121	1°32	87°0	0°082	BWE	50				

Table IV - Resolutions of polydisperse aerosols

Serial No.	Date	Production of Nuclei			Nuclei		Components of Aerosol			
		Time	Current	Duration	Age	Concentration	Diffusion Coefficient			
							D ⁱ	D ⁱⁱ	D ⁱⁱⁱ	D ^{iv}
		h	A	sec	h	Z	D ⁽ⁱ⁾ . 10 ⁶			
51	3- 8-61	10°3	4°90	60	2°3	21649	50	155		
52	20- 2-61	9°7	4°80	30	1°9	24542	50	170		
53	12- 5-61	15°1	4°90	30	1°6	43687	35	700		
54	2- 7-59	9°7	4°95	5	5°7	4400	7	45	250	
55	5- 4-60	9°5	4°75	45	5°8	7795	37	113	217	
56	17- 5-61	14°3	4°80	60	2°3	33300	40	150		
57	16- 8-61	9°7	4°90	30	2°5	16408	20	240		
58	27- 2-61	9°6	4°80	35	1°9	27114	33	100		
59	15- 5-61	10°9	4°85	60	1°3	56792	20	280	1000	
60	19- 1-62	9°6	4°20	45	2°7	5618	10	230	780	
61	16- 2-61	9°5	4°75	30	1°4	41644	62.5			
62	4- 8-61	10°2	4°90	60	2°6	30162	40	125		
63	4- 4-60	10°9	4°80	60	5°1	9670	32	108	600	
64	16- 2-61	9°5	4°75	30	1°9	30162	58			
65	28- 7-61	11°0	4°80	90	4°0	9105	35	110		
66	2- 5-60	9°5	4°80	90	6°0	7482	30	73	230	
67	26- 7-61	12°1	4°85	60	2°7	27661	30	100		
68	16- 5-61	14°6	4°90	90	1°9	41644	30	100		
69	4- 5-60	9°5	4°75	120	5°2	8456	15	45	150	
70	29- 9-61	-36	4°90	60	47	185	6	30	85	
71	1- 9-61	9°7	4°85	150	2°5	38328	15	33	100	
72	1- 8-61	10°8	4°90	150	4°6	13196	17	53		
73	9- 8-61	14°3	4°85	60	2°4	44674	15	50		
74	20-10-59	-7	6°10	5	19°5	650	5.5	36	147	500
75	25- 8-61	9°6	4°90	150	2°2	50776	10.5	33		

produced and stored in large gasometer (continued).

Components of Aerosol				Average		Diff. Coeff. to Average Radius	Z_v / Z	Diffusion Battery	Serial No.
Proportion				Diffusion Coefficient	Radius				
p'	p''	p'''	p''''	$\bar{D} \cdot 10^6$	$\bar{r} \cdot 10^6$	$D_r \cdot 10^6$			
0°437	0°563			109	1°32	87°0	0°073	BWE	51
0°512	0°488			119	1°36	82°3	0°085	BWE	52
0°550	0°450			334	1°38	80	0°277	BWE	53
0°055	0°347	0°598		165	1°39	79	0°011	III	54
0°300	0°573	0°127		103	1°39	79	0°010	BWE	55
0°396	0°604			106	1°39	79°0	0°092	BWE	56
0°325	0°675			169	1°46	71°8	0°149	BWE	57
0°276	0°724			81°5	1°50	68°1	0°015	BWE	58
0°395	0°417	0°188		313	1°51	67°2	0°033	BWE	59
0°255	0°317	0°428		410	1°51	67°2	0°166	BWE	60
1°000				62°5	1°57	62°5	0°145	BWE	61
0°540	0°460			79°1	1°58	61°5	0°126	BWE	62
0°425	0°525	0°050		100	1°60	60°2	0°023	BWE	63
1°000				58	1°63	58	0°163	BWE	64
0°480	0°520			74°0	1°63	58°0	0°132	BWE	65
0°328	0°572	0°100		74°6	1°67	55°5	0°020	BWE	66
0°420	0°580			70°6	1°68	54°7	0°137	BWE	67
0°509	0°491			64°4	1°78	49°2	0°167	BWE	68
0°108	0°722	0°170		59°6	1°88	44°4	0°007	BWE	69
0°118	0°315	0°567		58°4	2°15	34°4	0°039	BWE	70
0°284	0°435	0°281		46°7	2°26	31°3	0°020	BWE	71
0°551	0°449			33°2	2°50	25°9	0°118	BWE	72
0°486	0°514			33°0	2°54	25°2	0°123	BWE	73
0°231	0°450	0°241	0°078	91°9	2°58	24°4	0°061	III	74
0°256	0°744			27°2	2°68	22°8	0°039	BWE	75

Table IV - Resolutions of polydisperse aerosols

Serial No.	Date	Production of Nuclei			Nuclei		Components of Aerosol			
		Time	Current	Duration	Age	Concentration	Diffusion Coefficient			
							D'	D''	D'''	D''''
		h	A	sec	h	Z	$D^{(i)} \cdot 10^6$			
76	18- 5-61	14°7	4°80	180	1°9	56352	14	45		
77	31- 7-61	11°4	4°85	180	3°7	18728	15	90		
78	28- 4-60	-6°9	4°80	180	18	1900	15	22	80	
79	22-10-59	-7	7°00	4	20	800	5°0	42	100	
80	15- 8-61	9°7	4°90	180	2°2	58612	11	35		
81	28- 4-60	-6°9	4°80	180	18	2000	10°5	53	125	
82	6- 4-60	-7	4°75	180	23	2820	12°8	57		
83	19-10-59	-36	-	5	48	140	4°7	27	84	
84	10- 8-61	9°8	4°85	180	2°4	56792	6	20		
85	23-10-59	-31	7°00	4	43	180	5°0	54	100	
86	26-10-59	-36	4°80	30	48	470	4°5	35		
87	21-10-59	-31	6°10	5	43	90	4°0	38°5	173	
88	29-10-59	-35	4°80	60	47	280	4	20	70	
89	27-10-59	-60	4°80	30	72	80	4	15	60	
90	30-10-59	-59	4°80	60	71	105	3°8	27	90	
91	24- 5-60	-7	4°75	300	22	6000	2	7°5	15	50

produced and stored in large gasometer (concluded).

Components of Aerosol				Average		Diff. Coeff. to Average Radius	Min. Z_v/Z	Diffusion Battery	Serial No.				
Proportion				Diffusion Coefficient	Radius								
p ¹	p ²	p ³	p ⁴										
0°525	0°475			$\bar{D} \cdot 10^6$	$\bar{r} \cdot 10^6$	$D - \frac{r}{r} \cdot 10^6$							
0°716	0°284			28°7	2°72	22°1	0°145	B WE	76				
0°700	0°160	0°140		36°3	2°78	21°3	0°391	B WE	77				
0°290	0°620	0°090		25°2	2°98	18°8	0°223	B WE	78				
0°680	0°320			36°5	3°13	17°1	0°087	III	79				
0°750	0°170	0°080		18°7	3°39	14°8	0°095	B WE	80				
0°900	0°100			26°9	3°45	14°4	0°317	III	81				
0°336	0°298	0°366		17°2	3°46	14°2	0°261	B WE	82				
0°356	0°644			40°4	3°57	13°4	0°107	III	83				
0°534	0°279	0°187		15°0	3°85	11°7	0°116	B WE	84				
0°467	0°533			36°4	4°05	10°7	0°160	III	85				
0°470	0°460	0°070		20°8	4°26	9°8	0°159	III	86				
0°568	0°173	0°259		31°7	4°37	9°3	0°174	III	87				
0°654	0°140	0°206		23°9	4°96	7°5	0°212	III	88				
0°760	0°193	0°047		17°1	5°50	6°3	0°245	III	89				
0°700	0°179	0°038	0°065	12°3	6°13	5°2	0°295	III	90				
				10	8°90	2°8	0°262	III	91				

Serial No.	Analyst	D'	D''	D'''	p'	p''	p'''	\bar{r}
79	POLIAK	5°0	42	100	0°290	0°620	0°090	3°13
	METNIEKS	5°4	37°5	200	0°307	0°540	0°153	3°08
85	POLIAK	5°0	54	100	0°534	0°279	0°187	4°05
	METNIEKS	5°0	20	100	0°510	0°134	0°356	4°02

The sensitivity of the Exhaustion Method is remarkable. The average radius (\bar{r}) of the aerosol increases regularly when the age of the aerosol increases by as little as one hour. As an example, see the resolutions on 28 April 1961 (Serial Nos. 3, 4 & 5).

When the age of this aerosol increases from $5^{\circ}6$ to $6^{\circ}3$ and to $6^{\circ}9$ hrs, the average radius grows from $0^{\circ}56$ to $0^{\circ}58$ and to $0^{\circ}61 \cdot 10^{-6}$ cm.

Finally, Table IV enables one to study the composition (heterogeneity) of polydisperse aerosols of various modes of production, ages, etc. As an example we give in Fig.2 'point clouds' - well known from periodography - for $p^{(i)}$ vs $D^{(i)}$ of the aerosols listed in Table IV with average radius $\bar{r} < 1^{\circ}00 \cdot 10^{-6}$ and $> 2^{\circ}00 \cdot 10^{-6}$ cm.

There are 35 resolutions (Serial Nos. 1 to 35) of aerosols with $\bar{r} < 1^{\circ}00 \cdot 10^{-6}$ which have an average age of $2^{\circ}7$ hrs, and 22 (Serial Nos. 70 to 91) with $\bar{r} > 2^{\circ}00 \cdot 10^{-6}$ and an average age of $25^{\circ}5$ hrs. From Figs. 2a to c we deduce, in Table V, some characteristics of young and old aerosols stored in a large gasometer. Take e.g. Fig.2a, the cloud of points representing the component D' i.e. the component with the smallest diffusion coefficient or with the largest radius obtained by the Exhaustion Method for young and old

Table V - Extension of point clouds for components of young and old stored aerosols.

Aerosol	Young	Old
Average Age (hrs)	2°7	25°5
D*	$210 \cdot 10^{-6}$	$17 \cdot 10^{-6}$
D"	620	90
D'''	950	172
r*	$0^{\circ}84 \cdot 10^{-6}$	$3^{\circ}14 \cdot 10^{-6}$
r"	$0^{\circ}48$	$1^{\circ}30$
r'''	$0^{\circ}39$	$0^{\circ}93$
p*	47 %	90 %
p"	86	75
p'''	61	57

aerosols. In young aerosols we found D* values up to $210 \cdot 10^{-6}$ or components with radii as small as $0^{\circ}84 \cdot 10^{-6}$ cm, whereas in old aerosols the diffusion coefficients D* never exceeded $17 \cdot 10^{-6}$ or, the component with the largest radius was never smaller than $3^{\circ}14 \cdot 10^{-6}$ cm.

It appears, therefore, that the resolution of actual aerosols using the Exhaustion Method can supply - in addition to the average size of the aerosol nuclei - some information of the processes in an aging population of nuclei stored in a large gasometer.

REFERENCES

- (¹) L. W. POLLAK & A. L. METNIEKS: New calibration of photo-electric nucleus counters. *Geofisica Pura e Applicata*, Milano; Vol.43(1959), pp.285-301.
- (²) T. A. RICH and L. W. POLLAK & A. L. METNIEKS: Experiments with condensation nucleus size spectrometers. *Ibidem*; Vol.46(1960), pp.145-163.
- (³) L. W. POLLAK & A. L. METNIEKS: Extinction in a photo-electric nucleus counter using adiabatic expansion of 1:21 pressure ratio achieved by increasing or decreasing the original pressure. *Ibidem*; Vol.49 (1961), pp.208-216.
- (⁴) L. W. POLLAK & A. L. METNIEKS: On the validity of Boltzmann's law for small condensation nuclei. In the press.
- (⁵) L. W. POLLAK & A. L. METNIEKS: On the determination of the diffusion coefficient of heterogeneous aerosols by the dynamic method. *Geofisica Pura e Applicata*, Milano; Vol.37(1957), pp.183-190.
- (⁶) L. W. POLLAK, T. C. O'CONNOR & A. L. METNIEKS: On the determination of the diffusion coefficient of condensation nuclei using the static and dynamic method. *Ibidem*; Vol.34(1956), pp.177-195.
- (⁷) L. W. POLLAK, T. C. O'CONNOR, A. L. METNIEKS & R. FURTH: Report on the determination of the diffusion coefficient using the static and dynamic methods. *Ibidem*; Vol.36(1957), pp.70-75.
- (⁸) L. W. POLLAK & A. L. METNIEKS: The influence of air-flow on the determination of the diffusion coefficient of heterogeneous aerosols by the dynamic method. *Ibidem*; Vol.44(1959), pp.224-232.
- (⁹) J. J. NOLAN & V. H. GUERRINI: The diffusion coefficients and velocities of fall in air of atmospheric nuclei. *Proc. Roy. Irish Acad.*, Dublin; 43 A 2 (1935), pp.5-24.
- (¹⁰) J. J. NOLAN, P. J. NOLAN & P. G. GORMLEY: Diffusion and fall of atmospheric condensation nuclei. *Ibidem*; 45 A 4 (1938), pp.47-63.
- (¹¹) W. De MARCUS & J. W. THOMAS: Theory of a diffusion battery. Oak Ridge National Laboratory, Tennessee; Contract No. W-7405-eng-26 (1952).
- (¹²) J. W. THOMAS: The diffusion battery method for aerosol particle size determination. Health Physics Division, Oak Ridge National Laboratory, Tennessee; Contract No. W-7405-eng-26 (1953).

- (¹³) J. W. THOMAS: The diffusion battery method for aerosol particle size determination. *Journ. Colloid Science (U.S.A.)*; Vol.10(1955), pp.246-255.
- (¹⁴) P. J. NOLAN & J. DEIGNAN: Observations on atmospheric condensation nuclei in stored air. *Proc. Roy. Irish Acad., Dublin*; 51 A 18 (1948), pp.239-249.
- (¹⁵) V. CONRAD & L. W. POLLAK: Methods in Climatology. Harvard University Press, Cambridge, Mass. 1950.
- (¹⁶) A. L. METNIKES & L. W. POLLAK: Instruction for use of photo-electric condensation nucleus counters, their care and maintenance together with calibration and auxiliary tables. *Geophysical Bull. No.16 of the School of Cosmic Physics in the Dublin Institute for Advanced Studies; Dublin 1959.*
- (¹⁷) A. L. METNIKES & L. W. POLLAK: Tables and graphs for use in aerosol physics, Part I (Mobility v. Radius and vice versa). *Geophys. Bull. No.19 of the School of Cosmic Physics, Dublin; 1961.*
- (¹⁸) L. W. POLLAK & A. L. METNIKES: Intrinsic calibration of the photo-electric condensation nucleus counter Model 1957 with convergent light-beam. *Techn. (Scientific) Note No.9, Contract USAF 61(052) - 26; Dublin 1960.*
- (¹⁹) L. W. POLLAK & A. L. METNIKES: Photo-electric condensation nucleus counters of high precision for measuring low and very low concentrations of nuclei. *Geofisica Pura e Applicata, Milano; Vol.37(1957)*, pp. 174-182.
- (²⁰) L. W. POLLAK & J. DALY: A diffusion battery without end-pieces or connecting tubing. *Ibidem; Vol.45(1960)*, pp.249-257.

APPENDICES

APPENDIX I

A) Computation of 'observed' (apparent) diffusion coefficient as a function of air-flow.

Mixture of three aerosols

$$D^* = 20, \quad p^* = 0.333; \quad D'' = 100, \quad p'' = 0.333; \quad D''' = 200, \quad p''' = 0.333$$

$f = 46.25$ for $Q = 1$ litre/min (Diff. Batt. No.1 of School of Cosmic Physics, Dublin)

Q (litres/min)	fQ	D''/fQ	D'''/fQ	x''	x'''	Z_v''/Z_n''	Z_v'''/Z_n'''	Col. $5+6/7$	$\frac{1}{3} Col.8$	Z_v/Z	x	D 'Observed'										
												Column	1	2	3	4	5	6	7	8	9	10
0.2	9.250	2.162	-	-	0.1042	-	-	0.1042	0.0347	3.267	30.2											
0.5	23.125	0.865	4.324	8.650	0.3890	0.0121	0.0002	0.3953	0.1318	1.928	44.6											
1.0	46.250	0.432	2.162	4.324	0.5900	0.1042	0.0121	0.7063	0.2354	1.353	62.6											
1.5	69.375	0.288	1.441	2.883	0.6835	0.2164	0.0509	0.9508	0.3169	1.055	73.2											
2.0	92.500	0.216	1.081	2.162	0.7375	0.3087	0.1042	1.1504	0.3835	0.8635	79.9											
3.0	138.750	0.144	0.721	1.441	0.7990	0.4430	0.2164	1.4584	0.4861	0.6258	86.8											
4.0	185.000	0.108	0.541	1.081	0.8330	0.5300	0.3087	1.6717	0.5572	0.4898	90.6											
7.0	323.750	0.0618	0.309	0.618	0.8836	0.6690	0.4900	2.0426	0.6809	0.2921	94.6											
10.0	462.500	0.0432	0.216	0.432	0.9083	0.7375	0.5900	2.2358	0.7453	0.2064	95.5											

Notes to Appendix I.A :

Col.s. 2, 3, 4 from $D = x f Q$ (Eqn.4)

Col.s. 5, 6, 7 from tables for Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16)

taking figures in Col.s. 2, 3, 4 respectively as x

$$\text{Col.8 : } Z_v = \frac{Z^t}{V} + \frac{Z^n}{V} + \frac{Z^m}{V}$$

$$Z_v^t/Z^t + Z_v^n/Z^n + Z_v^m/Z^m = \frac{Z^t}{V}/p^t Z + \frac{Z^n}{V}/p^n Z + \frac{Z^m}{V}/p^m Z$$

$$p^t = p^n = p^m = 0.333 = 1/3$$

$$3\left(\frac{Z^t}{V} + \frac{Z^n}{V} + \frac{Z^m}{V}\right)/Z = 3 Z_v/Z$$

$$\text{Col.9 : Thus } Z_v/Z = \frac{1}{3} \left(Z_v^t/Z^t + Z_v^n/Z^n + Z_v^m/Z^m \right)$$

Col.10: from Col.9 using tables for Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16
(Ref.16)

Col.11: $D = x f Q$ (Eqn.4)

APPENDIX I

B*) Resolution of heterogeneous aerosol using the Exhaustion Method.

Mixture of three aerosols (Appendix I A)

$f = 46.25$ for $Q = 1$ litre/min (Diff. Batt. No.1 of School of Cosmic Physics, Dublin)

Q (litres/min)	D "Observed" (Apparent)	RQ	x	Z_v/Z	D'/fQ	Z_v'/Z'	$p'Z_v'/Z'$	Δ'	$\frac{\Delta'}{1-p'}$	Δ^* $\Delta^*/0.667$	x*	D*											
											Column	1	2	3	4	5	6	7	8	9	10	11	
0.2	30.2	9.250	3.267	0.0347	2.162	0.1042	0.0347	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
0.5	44.6	23.125	1.928	0.1318	0.8449	0.3890	0.1275	0.0043	0.0064	4.960	114.7												
1.0	62.6	46.250	1.353	0.2354	0.4324	0.5898	0.1964	0.0390	0.0585	2.745	127.0												
1.5	73.2	69.375	1.055	0.3169	0.2883	0.6893	0.2275	0.0894	0.1340	1.911	132.6												
2.0	79.9	92.500	0.8635	0.3835	0.2162	0.7374	0.2456	0.1379	0.2067	1.486	137.5												
3.0	86.8	138.750	0.6258	0.4861	0.1441	0.7989	0.2660	0.2201	0.3300	1.014	140.7												
4.0	90.6	185.000	0.4898	0.5572	0.1081	0.8329	0.2774	0.2798	0.4195	0.7755	143.5												
7.0	94.6	323.750	0.2921	0.6809	0.0618	0.8336	0.2942	0.3867	0.5798	0.4504	145.8												
10.0	95.5	462.500	0.2064	0.7453	0.0432	0.9083	0.3025	0.4428	0.6639	0.3162	146.2												

Notes to Appendix I B' :

(i) From graph of the D values in Col.1 (transferred from Appendix I A) we obtain by extrapolation for $Q = 0$: $D^* = \underline{20'0}$

(ii) With Eqn.6 $p^* = \exp(-D/fQ) : \exp(-D^*/fQ)$ taking for $Q = 0'2$, $f = 46'25$, $fQ = 9'250$,
 $D = 30'2$ (Col.1) $\underline{p^*} = \exp(-30'2/9'250) : \exp(-20'0/9'250) = \underline{0'333}$

(iii) Col.4: Z_v'/Z with x' in Col.3 using tables for Z_v'/Z vs x in Appendices III & IV of Geophys.

Bull. No.16 (Ref.16)

Col.6: Z_v'/Z with x' in Col.5 using tables Z_v'/Z vs x in Appendices III & IV of Geophys.

Bull. No.16 (Ref.16), taking x' as x

cols.7-11 according to Eqn.7

Col.8: $\Delta^* = \text{Col.4} - \text{Col.7}$

Col.10: x^* from Col.9 by treating $\Delta^*/(1 - p^*)$ as Z_v'/Z and using tables Z_v'/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16)

Col.11: $D^* = x^* fQ$

APPENDIX I

Bⁿ) Resolution of heterogeneous aerosol using the Exhaustion Method (Concluded).

Mixture of three aerosols (Appendix I A)

$f = 46.25$ for $Q = 1$ litre/min (Diff. Batt. No.1 of School of Cosmic Physics, Dublin)

Q (litres/min)	D^*	Δ^*	RQ	D^n / RQ	Z^n / Z_V^n	$p^n Z^n / Z^n$ $p^n = 0.341$	Δ^n	$1 - p^n Z^n / Z^n$ $\Delta^n / 0.326$	x^{**}	D^{**}
Column	1	2	3	4	5	6	7	8	9	10
0'2	-	-	-	-	-	-	-	-	-	-
0'5	114°7	0.0043	23°125	4°281	0°0126	0°0043	-	-	-	-
1°0	127°0	0.0390	46°250	2°141	0°1065	0°0363	0°0027	0°0083	4°700	217
1°5	132°6	0.0894	69°375	1°427	0°2194	0°0748	0°0146	0°0448	3°010	209
2°0	137°5	0.1379	92°500	1°070	0°3122	0°1065	0°0314	0°0963	2°243	207
3°0	140°7	0.2201	138°750	0°7135	0°4462	0°1522	0°0679	0°2083	1°479	205
4°0	143°5	0.2798	185°000	0°5351	0°5330	0°1818	0°0980	0°3006	1°107	205
7°0	145°8	0.3867	323°750	0°3058	0°6711	0°2288	0°1579	0°4844	0°6302	204
10'0	146°2	0°4428	462°500	0°2141	0°7389	0°2520	0°1908	0°5853	0°4404	204

Notes to Appendix I Bⁿ:

(i) D^* (Col.1) and Δ^* (Col.2) transferred from Appendix I B^f

(ii) From graph of the D^* values in Col.1 we obtain by extrapolation for $Q = 0$: $D^n = \underline{99}$

(iii) With Eqn.8 we compute $p^n = (1 - p^f) \exp(-D^n/fQ) : \exp(-D^*/fQ)$ taking for $Q = 0.5$,
 $f = 46.25$, $fQ = 23.125$; $D^* = 114.7$ (for $Q = 0.5$), $p^f = 0.333$, $1 - p^f = 0.667$ and
obtain $p^n = \underline{0.341}$

(iv) Col.5: Z_v^n/Z^n with x^n in Col.4 using tables for Z_v/Z vs x in Appendices III & IV of
Geophys. Bull. No.16 (Ref.16), taking x^n as x

Col.9: x^{**} from Col.8 by treating $\Delta^n/(1 - p^n - p^n)$ as Z_v/Z and using tables of
 Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16)

Col.10: $D^{**} = x^{**} f Q$

(v) From graph of the D^{**} values of Col.10 we obtain by extrapolation for $Q = 0$:
 $D^{**n} = \underline{207}$ (average of the D^{**})

APPENDIX II

A) Computation of 'observed' (apparent) diffusion coefficient as a function of air-flow.

Mixture of nine aerosols with Gaussian distribution of their concentration

Diffusion Battery BWE 1 (G.E., Schenectady); f (1 litre/min) = 58°67

x_1

D_1	60	100	140	180	220	260	300	340	380
$x_1 = D_1/fQ$	x_1	x_2	x_3	x_4	x_5	x_6	x_7	x_8	x_9
Q	fQ								
0°10	5°867	10°226	-	-	-	-	-	-	-
0°25	14°6675	4°091	6°818	9°545	-	-	-	-	-
0°50	29°335	2°045	3°409	4°772	6°136	7°500	8°863	10°227	-
1°00	58°67	1°023	1°704	2°386	3°068	3°750	4°432	5°113	5°795
2°50	146°675	0°409	0°682	0°954	1°227	1°500	1°773	2°045	6°477
5°00	293°35	0°205	0°341	0°477	0°614	0°750	0°886	1°023	2°318
10°00	586°7	0°1023	0°1704	0°239	0°307	0°375	0°443	1°159	2°591
20°00	1173°4	0°0511	0°0852	0°1193	0°1534	0°1875	0°2216	0°256	1°295
50°00	2933°5	0°0205	0°0341	0°0477	0°0614	0°0750	0°0886	0°1023	0°1159
									0°1295

Note: $D = x_1 f Q$, Q in litres/min

APPENDIX II A (Continued)

$(Z_v/Z)_1$

Q	1	2	3	4	5	6	7	8	9
0'10	$33 \cdot 10^{-6}$	-	-	-	-	-	-	-	-
0'25	0'0152	$994 \cdot 10^{-6}$	$64 \cdot 10^{-6}$	-	-	-	-	-	-
0'50	0'1175	0'0301	0'0077	$192 \cdot 10^{-5}$	$50 \cdot 10^{-5}$	$13 \cdot 10^{-5}$	$33 \cdot 10^{-6}$	-	-
1'00	0'3270	0'1658	0'0841	0'0423	0'0214	0'0108	0'0055	$276 \cdot 10^{-5}$	$140 \cdot 10^{-5}$
2'50	0'6045	0'4600	0'3500	0'2670	0'2038	0'1543	0'1175	0'0897	0'0685
5'00	0'7460	0'6475	0'5650	0'4920	0'4305	0'3750	0'3270	0'2855	0'2492
10'00	0'8387	0'7753	0'7190	0'6700	0'6255	0'5840	0'5460	0'5100	0'4760
20'00	0'8974	0'8562	0'8217	0'7896	0'7605	0'7327	0'7065	0'6820	0'6590
50'00	0'9438	0'9216	0'9020	0'8841	0'8679	0'8525	0'8387	0'8251	0'8125

Note: $(Z_v/Z)_1$ with x_i of previous table using tables for Z_v/Z versus x in Appendices III & IV in Geophysical Bull. No.16 (Ref.16)

APPENDIX III A (Concluded)

$$(Z_v)_i = (Z_v/Z)_i \cdot Z_i$$

Z_i	140	475	1170	2010	2410	2010	1170	475	140	$\Sigma(Z_v)_i$	x	D †Observed†
1	1	2	3	4	5	6	7	8	9			
Q												
0°10'	0°0046	-	-	-	-	-	-	-	-	0°0046	14°5	85°1
0°25'	2°13'	0°47'	0°07'	-	-	-	-	-	-	2°67	8°14	119°4
0°50'	16°45'	14°30'	9°01'	3°86'	1°21'	0°26'	0°04	-	-	45°13	5°306	155°7
1°00'	45°8	78°8	98°4	85°0	51°6	21°7	6°4	1°3	0°2	389°2	3°152	184°9
2°50'	84°6	218°5	409°5	536°7	491°2	310°1	137°5	42°6	9°6	2240°3	1°405	206°1
5°00'	104°4	307°6	661°1	988°9	1037°5	753°8	382°6	135°6	34°9	4406°4	0°7267	213°2
10°00'	117°4	366°3	841°2	1346°7	1507°5	1173°8	698°8	242°2	66°6	6302°5	0°3675	215°6
20°00'	125°6	406°7	961°4	1587°1	1832°8	1472°7	826°6	324°0	92°3	7629°2	0°1842	216°1
50°00'	132°1	437°8	1055°3	1777°0	2091°6	1713°5	981°3	391°9	113°8	8694°3	0°0736	215°9

Notes:

(1) $(Z_v)_i$ is obtained by multiplying the $(Z_v/Z)_i$ in the $(i+1)$ th column of the previous table with the Z_i shown in the heading of the $(i+1)$ th column of this table

(11) The x in the last but one column is obtained from $\Sigma(Z_v)_i / Z$, $Z = 10000$, using tables for Z_v/Z vs x in Appendices III & IV of Geophysical Bull. No.16 (Ref.16)

(iii) D (last column) = $x f Q$

APPENDIX II

B*) Resolution of heterogeneous aerosol using the Exhaustion Method.

Mixture of nine aerosols with Gaussian distribution of their concentration (Appendix II A)

Diffusion Battery BWE 1 (G.E., Schenectady); f (1 litre/min) = 58°67

Q (litres/min)	D	*Observed* (Apparent)	fQ	x	Z_v/Z	D/fQ	$D = 62$	Z_v^*/Z^*	$p^*Z_v^*/Z^*$ $p^* = 0^*020$	Δ^*	$\frac{\Delta^*}{1-p^*}$ $1-p^* = 0^*980$	x^*	D^*
Column	1	2	3	4	5	6	7	8	9	10	11		
0°10	85°1	5°867	14°5	$46 \cdot 10^{-8}$	10°57	$234 \cdot 10^{-7}$	$4668 \cdot 10^{-9}$	-	-	-	-		
0°25	119°4	14°6675	8°14	$267 \cdot 10^{-6}$	4°227	0^*01326	$265 \cdot 10^{-6}$	-	-	-	-		
0°50	155°7	29°335	5°306	0^*00451	2°114	0^*1095	0^*00219	0^*00232	0^*00237	5^*95	174^*5		
1°00	184°9	58°67	3°152	0^*03892	1°057	0^*3163	0^*00633	0^*03259	0^*03326	3^*309	194^*1		
2°50	206°1	146°675	1°405	0°2240	0°4227	0^*5957	0^*0119	0^*2121	0^*2164	1^*441	211^*4		
5°00	213°2	293°35	0°7267	0°4406	0°2114	0^*7413	0^*0148	0^*4258	0^*4345	0^*741	217^*4		
10°00	215°6	586°7	0°3675	0°6902	0°1057	0^*8353	0^*0167	0^*6135	0^*6260	0^*374	219^*4		
20°00	216°1	1173°4	0°1842	0°7629	0°05284	0^*8951	0^*0179	0^*7450	0^*7602	0^*1878	220^*4		
50°00	215°9	2933°5	0°0736	0°8694	0°02114	0^*9427	0^*0189	0^*8505	0^*8679	0^*0750	220^*0		

Notes to Appendix II B*

(i) From graph of the D values in Col.1 (transferred from Appendix II A) we obtain by extrapolation for $Q = 0$: $D^* = \underline{62}$

(ii) With Equ.6 $p^* = \exp(-D/fQ) : \exp(-D/fQ)$ taking for $Q = 0^*10$, $f = 58^*67$,
 $fQ = 5^*867$, $D = 85^*1$ (Col.1) $\underline{p^*} = \exp(-85^*1/5^*867) : \exp(-62/5^*867) = 0^*020$

(iii) Col.4: Z_v/Z with x in Col.3 using tables for Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16)

Col.6: Z_v^*/Z^* with x^* in Col.5 using tables Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16), taking x^* as x

cols.7 - 11 according to Equ.7

Col.8: $\Delta^* = \text{Col.4} - \text{Col.7}$

Col.10: x^* from Col.9 by treating $\Delta^*/(1 - p^*)$ as Z_v/Z and using tables Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16)

Col.11: $D^* = x^* f Q$

APPENDIX II

Bⁿ) Resolution of heterogeneous aerosol using the Exhaustion Method (Concluded).

Mixture of nine aerosols with Gaussian distribution of their concentration (Appendix II A)

Diffusion Battery BWE 1 (G.E., Schenectady); f (1 litre/min) = 58°67

Q (litres/min)	D*	Δ^*	fQ	$D^n / \Delta Q$ $D^n = 155$	Z^n / Z^n $p^n = 0.503$	$p^n Z^n / Z^n$ $\Delta^n - \frac{p^n Z^n}{Z^n} / 0.477$	Δ^n		$1 - p^n - p^n$ $\Delta^n / 0.477$	χ^{**}	D**
							Column	1	2	3	4
0.5	174.5	0.00232	29.335	5.284	0.00461	0.00232	-	-	-	-	-
1.0	194.1	0.03259	58.67	2.642	0.0648	0.03259	-	-	-	-	-
2.5	211.4	0.2121	146.675	1.057	0.3163	0.1591	0.0530	0.1111	2.10	308.0	
5.0	217.4	0.4258	293.35	0.5284	0.5368	0.2700	0.1558	0.3266	1.024	300.4	
10.0	219.4	0.6135	586.7	0.2642	0.7008	0.325	0.2610	0.5472	0.5086	298.4	
20.0	220.4	0.7450	1173.4	0.1321	0.8099	0.4074	0.3376	0.7078	0.2542	298.3	
50.0	220.0	0.8505	2933.5	0.0528	0.8951	0.4502	0.4003	0.8392	0.1018	298.6	

Notes to Appendix II Bⁿ :

(1) D* (Col.1) and Δ* (Col.2) transferred from Appendix II B^t

(11) From graph of the D* values in Col.1 we obtain by extrapolation for Q = 0 :

$$\underline{D^n = 155}$$

(11.1) With Eqn.8 we compute $p^n = (1 - p^t) \exp(-D^n/fQ) : \exp(-D^n/fQ)$ taking for $Q = 0'5$,
 $f = 58'67$, $fQ = 29'335$; $D^* = 174'5$ (for $Q = 0'5$), $D^n = 155$, $p^t = 0'020$,
 $1 - p^t = 0'980$ and obtain $\underline{p^n = 0'503}$

(14) Col.5: Z_v^n/Z_v^n with x^n in Col.4 using tables for Z_v/Z vs x in Appendices III & IV
of Geophys. Bull. No.16 (Ref.16), taking x^n as x
Col.9: x^{**} from Col.8 by treating $\Delta^n/(1 - p^t - p^n)$ as Z_v/Z and using tables of
 Z_v/Z vs x in Appendices III & IV of Geophys. Bull. No.16 (Ref.16)
Col.10: $D^{**} = \underline{x^{**} f Q}$

(v) From graph of the D** values of Col.10 we obtain by extrapolation for Q = 0 :

$$\underline{D^{**} = 201} \text{ (Average of the } D^{**})$$

APPENDIX III

A) Measurements for resolution of heterogeneous aerosol
using the Exhaustion Method on 21 January 1962, interpolated for 11h 53m.

Artificially produced aerosol stored in large gasometer

Diffusion Battery BWE 1 (G.E., Schenectady), $f(1 \text{ litre/min}) = 58^\circ 67$;
Counter of BWE 1 with ceramic lining and convergent light-beam

Column	Q (litres/min)	Time	Type	Interpol. Counter Reading	Z	Z_v	Z_v/Z	x	r_Q	D	$D = x r_Q$
20'0	11 53	2	Z_v	49'5	11492						
0'5	11 53		Z_v	92'3		215	0'0187	3'885	29'335	113'966	
1'0	11 53		Z_v	89'5		340	0'0296	3'427	58'670	201'062	
2'5	11 53		Z_v	78'5		1241	0'1080	2'128	146'675	312'124	
5'0	11 53		Z_v	69'1		2854	0'248	1'300	293'350	381'355	
10'0	11 53		Z_v	61'1		5194	0'452	0'700	586'700	410'690	
20'0	11 53		Z_v	56'2		7330	0'638	0'355	1173'400	416'557	

Col.4 & 5: From 1960 Calibration Table in Ref.18

Col.7 : From Appendices III and IV of Geophys. Bull. No.16 (Ref.16)

Col.9 : Three decimals for plotting superfluous, but x in Col.7
to three decimals has to be transferred to Appendix III B*

APPENDIX III

B¹) Resolution of heterogeneous aerosol using the Exhaustion Method.

Artificially produced aerosol stored in large gasometer

Measurements on 21 January 1962, interpolated for 11h 53m (Appendix III A)

Diffusion Battery BWE 1 (G.E., Schenectady); f (1 litre/min) = 58°67

Q (litres/min)	D	fQ	x	Z_V / fQ	Z_V^* / fQ	$p^* Z_V^* / Z_V$	Δ^*	$\frac{\Delta^*}{1 - p^*}$	x^*	D*
				$D^* = 26$		$p^* = 0.050$		$1 - p^* = 0.95$		
Column	1	2	3	4	5	6	7	8	9	10
0°5	113°966	29°335	3°885	0°0187	0°8863	0°3749	0°0187	-	-	-
1°0	201°062	58°670	3°427	0°0296	0°4432	0°5839	0°0292	-	-	-
2°5	312°124	146°675	2°128	0°1080	0°1773	0°7687	0°0384	0°0696	0°0733	2°528 371
5°0	381°355	293°350	1°300	0°248	0°0886	0°8525	0°0426	0°2054	0°2162	1°442 423
10°0	410°690	586°700	0°700	0°452	0°0443	0°907	0°0454	0°4066	0°4280	0°756 444
20°0	416°557	1173°400	0°355	0°638	0°0222	0°941	0°0471	0°5909	0°6220	0°381 447

Notes to Appendix III B* :

(1) From graph of the D values in Col.1 (transferred from Appendix III A) we obtain by extrapolation for $Q = 0$: $D^* = 26$

(11) With Equ.6 $p^* = \exp(-D^*/fQ) : \exp(-D^*/fQ)$ taking for $Q = 0^*5$, $f = 58^*67$,

$$fQ = 29^*335, D = 113^*966 \text{ (Col.1)} \quad p^* = \underline{\exp(-113^*966/29^*335)} : \exp(-26/29^*335) = \underline{0^*050}$$

(111) Col.4: Z_v/Z with x in Col.3 using tables for Z_v/Z vs x in Appendices III and IV of Geophys. Bull. No.16 (Ref.16)

Col.6: Z_v^*/Z^* with x^* in Col.5 using tables Z_v/Z vs x in Appendices III and IV of Geophys. Bull. No.16 (Ref.16)

cols.7-11 according to Equ.7

Col.8: $\Delta^* = \text{Col.4} - \text{Col.7}$

Col.10: x^* from Col.9 by treating $\Delta^*/(1 - p^*)$ as Z_v/Z and using tables Z_v/Z vs x in Appendices III and IV of Geophys. Bull. No.16 (Ref.16)

Col.11: $D^* = x^* f Q$

APPENDIX III

Bⁿ) Resolution of heterogeneous aerosol using the Exhaustion Method (Concluded).

Artificially produced aerosol stored in large gasometer

Measurements on 21 January 1962, interpolated for 11h 53m (Appendix III A)

Diffusion Battery BWE 1 (G.E., Schenectady); f (1 litre/min) = 58°67

Q (litres/min)	D*	Δ^*	RQ	D^n / fQ $D^n = 300$	Z^n_V / Z^n	$P^n Z^n / Z^n$ $P^n = 0.592$	Δ^n	$\frac{\Delta^n}{1 - P^n}$	x**	D**
							$P^n Z^n / Z^n$ $\Delta^n / 0.358$			
Column	1	2	3	4	5	6	7	8	9	10
2.5	371	0.0696	146.675	2.045	0.1175	0.0696	-	-	-	-
5.0	423	0.2054	293.350	1.023	0.3270	0.1936	0.0118	0.0330	3.320	974
10.0	444	0.4066	586.700	0.511	0.5460	0.3232	0.0894	0.2330	1.364	800
20.0	447	0.5909	1173.400	0.2557	0.7067	0.4184	0.1725	0.4818	0.6354	746

Notes to Appendix III Bⁿ :

(i) D* (Col.1) and Δ* (Col.2) transferred from Appendix III Bⁿ

(ii) From graph of the D* values in Col.1 we obtain by extrapolation for Q = 0 :

$$\underline{D^n = 300}$$

(iii) With Equ.8 we compute $p^n = (1 - p^*) \exp(-D^n/fQ) : \exp(-D^n/fQ)$ taking for $Q = 2^{\circ}5'$,

$$f = 58^{\circ}67, fQ = 146^{\circ}675; D^* = 371 \text{ (for } Q = 0^{\circ}5\text{)}, D^n = 300, p^* = 0^{\circ}050,$$

$$1 - p^* = 0^{\circ}950 \text{ and obtain } \underline{p^n = 0^{\circ}592}$$

(iv) Col.5: Z_v^n/Z^n with xⁿ in Col.4 using tables for Z_v/Z vs x in Appendices III & IV

of Geophys. Bull. No.16 (Ref.16), taking xⁿ as x

Col.9: x^{**} from Col.8 by treating $\Delta^n/(1 - p^* - p^n)$ as Z_v/Z and using tables of Z_v/Z vs x in Appendices III and IV of Geophys. Bull. No.16 (Ref.16)

$$\text{Col.10: } D^{**} = x^{**} fQ$$

(v) From graph of the D^{**} values of Col.10 we obtain by extrapolation for Q = 0 :

$$\underline{D^{**} = 240 \text{ (Average of the } D^{**})}$$

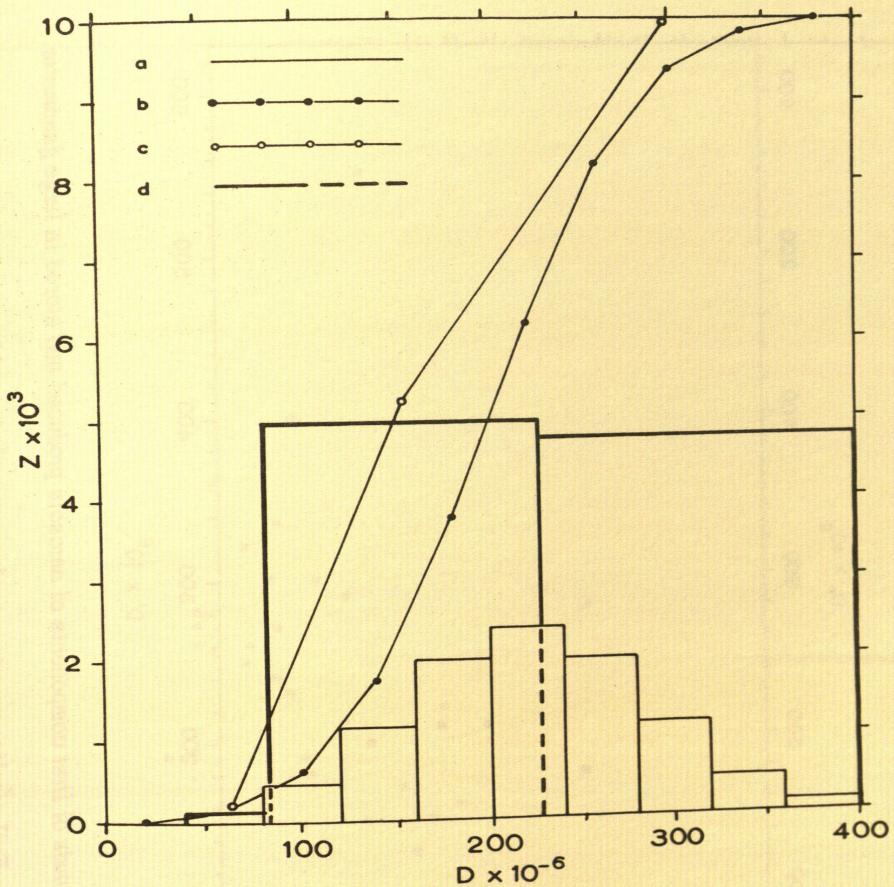


Fig. 1 - Heterogeneous aerosol containing nine components with Gaussian distribution of their concentration.

- a *Histogram of original Z_i vs D_i (Table 1), b Ogive of Z_i ,*
- c *Ogive of the three components obtained by Exhaustion Method,*
- d *Histogram of the three components obtained by Exhaustion Method*

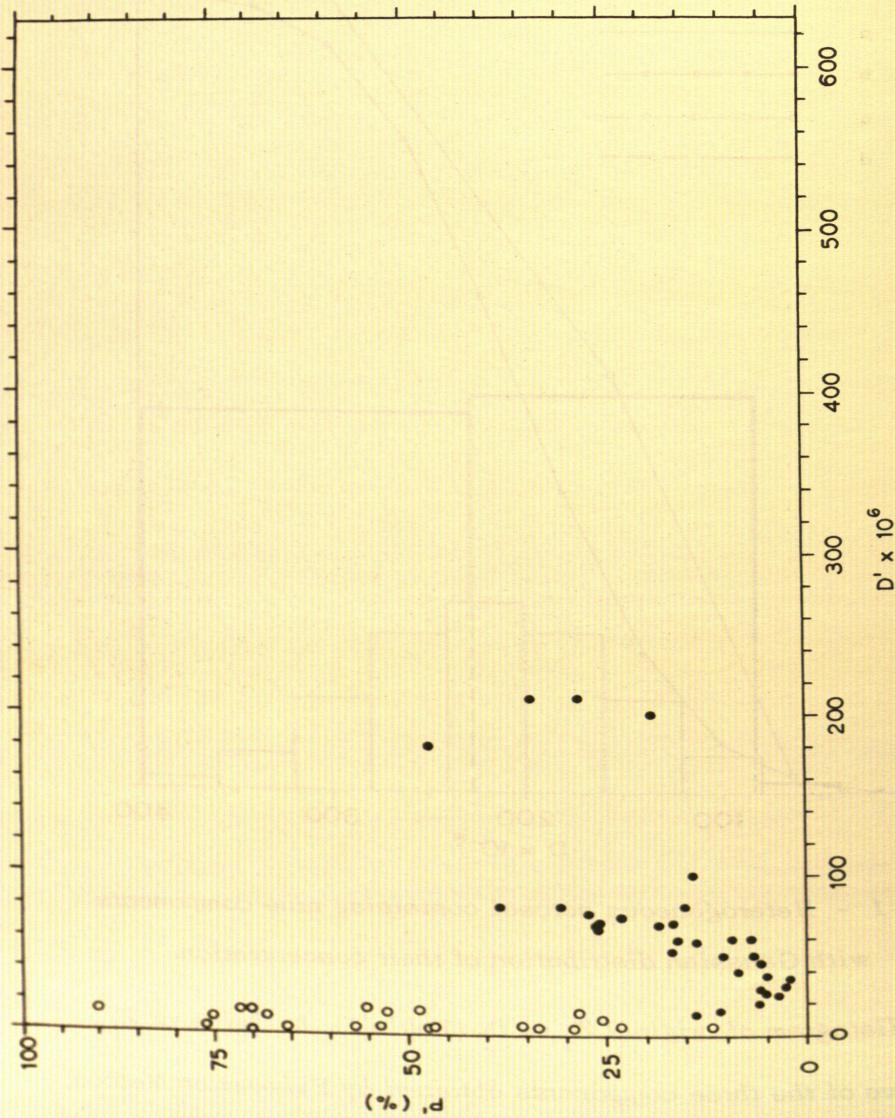


Fig. 2a - Point clouds of first components of aerosols produced and stored in large gassometer.

Full circles: young aerosols; open circles: old aerosols

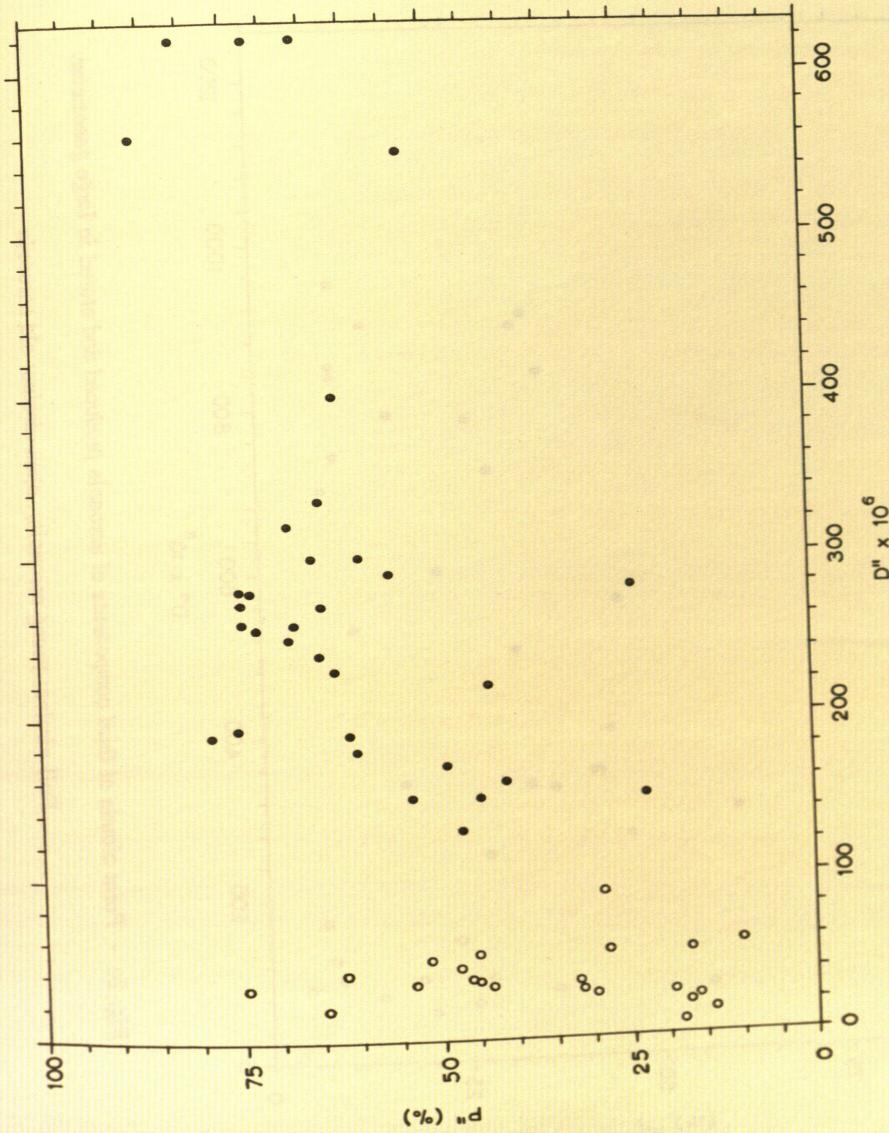


Fig. 2b - Point clouds of second components of aerosols produced and stored in large gasometer.

Full circles: young aerosols; open circles: old aerosols

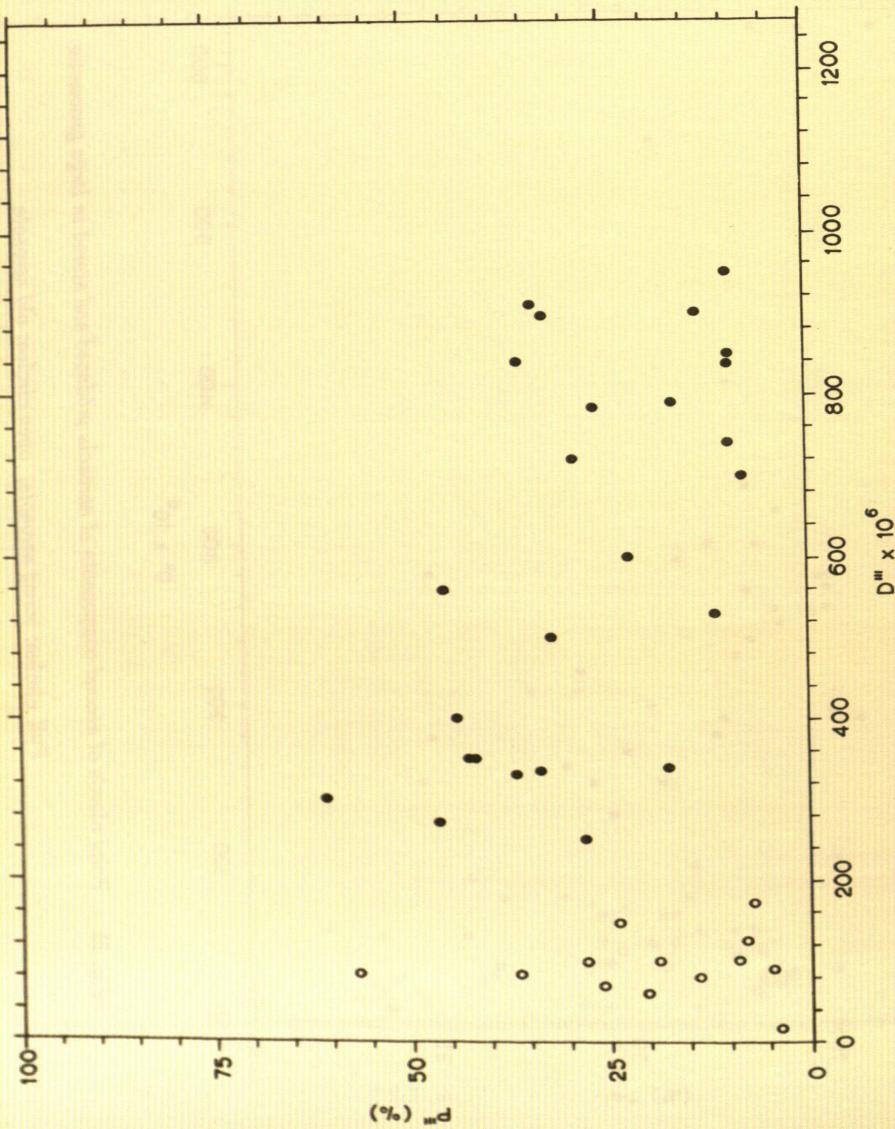


Fig. 2c - Point clouds of third components of aerosols produced and stored in large gasometer.

Full circles: young aerosols; open circles: old aerosols