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Studies on colloidal particles; scavenging of aerosol
particles by a falling macroscopic particle

Annual Progress Report

Milton Kerker

Clarkson College of Technology
Potsdam, New York

January 1, 1972 - December 31, 1972

Prepared for the U.S. Atomic Energy Commission
Under Contract No. AT(11-1)-3257

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ABSTRACT

Measurements of the collection efficiency of a freely falling water droplet falling through a silver chloride aerosol have been carried out. The apparatus was modified to permit preparation of the aerosol in nitrogen rather than helium (in order to simulate air) and to vary the distance over which the droplet accelerates prior to entry into the scavenging chamber from 30 cm up to 515 cm.

The effect of particle size was studied. For a 0.158 cm droplet and accelerating distance of 30 cm, particle radius ranged from 0.16 to 0.46 μm . For a 0.254 cm droplet and accelerating distance of 515 cm, particle radius varied from 0.12 to 0.36 μm . The results were correlated with Péclet number using a newly developed theory assuming potential flow and diffusion through a concentration boundary layer.

For a particle radius 0.36 μm , the effect of Reynold's number was studied for droplet radii 0.116, 0.158, and 0.254 cm

and for acceleration distances 30 cm, 298 cm and 515 cm. A very strong dependence upon Reynold's number was observed. These results were extrapolated to give collection efficiencies at terminal velocity for four droplet sizes. With additional results to be obtained during the coming period, calculations will be carried out of the rate of washout of submicron aerosol with rainfall rate as well as the effect upon atmospheric visibility.

I Introduction

Note: The principal investigator has devoted about 25% of his time to this project during the three summer months (June, July, August 1972) and about 10% of his time during the academic year (May, Sept - Jan 1972). His effort will continue at the same level for the remainder of the contract period.

The problem under investigation is to measure the collection efficiency of a water droplet falling through an aerosol consisting of submicron particles. The goals are at least twofold:

1. To calculate the rate of scavenging of the atmospheric aerosol by rain in order to determine its effect upon transport of the aerosol to the ground as well as upon atmospheric visibility.

2. To explicate theoretically the mechanism of capture.

A great deal of data have been obtained this year, and these bring the goals much closer. This progress report comes just at a time when there has been a surge of results and there has hardly been time to develop these results in a form suitable for journal publication by the 31 January deadline. This is our apology

for presenting them in such crude form in this progress report. Also, as will be apparent from our discussion, there are still additional data which must be obtained.

The experiment consists in dropping a water droplet of radius A through a silver chloride aerosol consisting of a narrow distribution of spherical particles of modal radius a_M and a breadth parameter σ_0 . The droplet accelerates for a distance S before entering the scavenging chamber containing the aerosol. The apparatus is depicted schematically in Fig. 1. The aerosol is prepared in a three-stage thermal generator; the size distribution of each aerosol sample is determined by light scattering; the mass concentration is determined by weighing after collection either by thermal precipitation or filtration; the silver chloride captured by the falling droplet is analyzed by a very sensitive colorimetric technique. Each experimental run is quite long and tedious because of (1) the long warm-up period required for stable operation of the aerosol generator, (2) the difficulty in aiming the water droplet so that it falls into the collection device, (3) the large amount of data and computing necessary for determination of the particle size distribution by light scattering, and (4) the requirement that for each run a large number of droplets be utilized. The latter requirement is a consequence of the very small amounts of aerosol collected by each droplet.

In the first stage of this work, helium was the carrier gas for the aerosol. We have generally found such aerosols easier to work with, presumably because of the high thermal conductivity of helium. The earlier results were all carried out with an

accelerating distance $S = 30$ cm, and the effect of droplet size and particle size was studied. The results were consistent with a mechanism of collection by Brownian diffusion of the particles to the droplet as evidenced by their being correlated by the Péclet number, but they differed quantitatively from the expression based upon creeping flow developed by Friedlander (1) and Zebel (2). (Technical Progress Reports COO-3257, Reports No. 1 and 2. A reprint of the latter is enclosed with this report.)

The last progress report described the work involved in the preparation of silver chloride aerosols in nitrogen rather than in helium. Nitrogen was selected in order to correspond more closely to conditions in the atmosphere. These new aerosols have been utilized to study the effect of particle size, droplet size, and Reynold's number upon the collection efficiency.

II Theoretical interpretation of dependence of collection efficiency upon particle size for the 30 cm acceleration tube

Professor S.K. Suneja, who recently joined Clarkson's Chemical Engineering Department, has collaborated with us in the theoretical interpretation of the results published in COO-3257, Report No. 2 (4). A recent paper (5), accepted for publication in the Journal of Colloid and Interface Science, is appended as COO-3257, Report No. 5. The treatment differs from the earlier creeping flow treatment of Friedlander and Zebel by assuming that the droplet is falling at high speed with a consequent neglect of the viscous terms in the momentum equations. The capture is treated as occurring due to the diffusion of the particles through a concentration boundary layer around the droplet. According to this theory, the collection efficiency is given by

$$E = 2.18 \text{ Pe}^{-1/2} + 2I \text{ for } I \leq 0.3 \text{ Pe}^{-1/2}$$

compared to $3.18 \text{ Pe}^{-2/3}$ for slowly falling drops where the Péclet number $\text{Pe} = A u_{\infty}/D$, and $I = a/A$ with u_{∞} the velocity relative to the bulk of the fluid and D is the particle diffusion coefficient. This treatment provides much better agreement with the data and is certainly more realistic than the creeping flow approximation.

Professor Suneja is now extending the theory to include finite Reynold's number as well as the effects of the wake in order to understand the newer data which are described below.

III Survey of experiments

Our laboratory was moved this year to a new building occupied by the Chemistry, Physics, and Mathematics Departments. This provided the opportunity of increasing the height of the apparatus by extending through two floors. It is now possible to vary the acceleration distance from 30 cm as used earlier up to 515 cm. In Table I we have listed the velocities, Reynold's number, and percentage of terminal velocities attained in nitrogen by three drop sizes ($A = 0.116, 0.158, \text{ and } 0.254 \text{ cm}$) upon entrance to and upon exit from the 135 cm scavenging tube after dropping through three acceleration tubes ($S = 30, 298 \text{ and } 515 \text{ cm}$). Values for the 0.116 cm droplet in helium for 30 cm tube are also included.

Figure 2 depicts the domain representing the combinations of acceleration distances and droplet sizes. The x-markings represent values for which experiments have been carried out this year. The modal particle radius for these averaged $0.36 \text{ }\mu\text{m}$ with a standard deviation of $0.02 \text{ }\mu\text{m}$. In addition, the effect of particle size

was studied for $A = 0.158$ cm, $S = 30$ cm, and for $A = 0.254$ cm, $S = 515$ cm over the particle size ranges 0.16 to 0.46 μm and 0.12 to 0.36 μm , respectively. The earlier particle size study (Technical Progress Report COO-3257, Report No. 2) in helium was for $A = 0.254$ cm, $S = 30$ cm for $a_M = 0.20$ to 0.52 μm . The Δ -markings represent values for which experiments are planned, including those encircled for which the particle size effect will also be studied.

IV Effect of particle size

The results of the studies of the effect of particle size are shown in Tables II and III and are plotted in Figs. 3-6. These results are also plotted against Péclet number on Fig. 7 along with earlier data obtained in helium. The least squares straight line fit to these data yields for $A = 0.254$ cm, $S = 515$ cm,

$$E = 3.02 \text{ Pé}^{-0.47} \quad (1)$$

for $A = 0.254$ cm, $S = 30$ cm (in helium)

$$E = 4.10 \times 10^4 \text{ Pé}^{-1.01} \quad (2)$$

and for $A = 0.158$ cm, $S = 30$ cm

$$E = 1.39 \times 10^8 \text{ Pé}^{-1.51} \quad (3)$$

with correlation coefficients of 0.968, 0.988, and 0.884, respectively.

Alternatively, they can be compared with curves shown in Fig. 7 for Zebel's theory for creeping flow

$$E = 3.18 \text{ Pé}^{-2/3} \text{ for } I = 0 \quad (4)$$

and with Suneja's theory for potential flow

$$E = 2.18 \text{ Pé}^{-1/2} + 2I \text{ for } I \leq 0.3 \text{ Pé}^{-1/2} \quad (5)$$

where I is the ratio of the particle and droplet diameter.

There is good agreement with Suneja's theory for the 0.254 cm droplet despite the finite values of the Reynold's number and the large value of I compared with $0.3 \text{ Pé}^{-1/2}$. The efficiency for the 0.158 cm droplet falls between the two limiting cases, as one might expect considering the lower Reynold's number. However, the slope is greater than predicted theoretically.

It is now apparent that in order to better elucidate the dependence of the collection efficiency upon aerosol particle size, particularly in the neighborhood of the terminal velocity, it will be necessary to obtain further data as indicated in Fig. 2.

V. Effect of Reynold's number

The results for three droplet sizes ($A = 0.116, 0.158, \text{ and } 0.254 \text{ cm}$) are presented in Tables IV, V, and VI for each of three accelerating distances ($S = 30, 298, \text{ and } 515 \text{ cm}$). The corresponding Reynold's numbers, Péclet numbers, aerosol particle size, and values of I are also given.

These results are plotted versus particle radius in Fig. 8. Figures 9 and 10 show the same data plotted against Reynold's and Péclet numbers, respectively, with the droplet radii as parameters.

Figure 9 provides the most illuminating way of viewing these results which expose primarily the influence of the fluid mechanics of the droplet just as Fig. 7 exposes primarily the influence of particle size. The most striking aspect of the results for $A = 0.116 \text{ cm}$ and $A = 0.158 \text{ cm}$ is the very sharp

dependence of the collection efficiency upon the Reynold's number. This exceeds by far anything that we have been able to account for in terms of the usual model of Brownian diffusion through the concentration boundary layer even taking into account the possibility of flattening of the droplet or of the Magnus effect.

There are two other possibilities. One of these is the additional collection on the backside due to the effect of the wake. The other is the possibility of collection by the inertial mechanism. The Stokes number at the high velocities approaches the value at which inertial collection becomes significant ($St = 0.1$). Both of these aspects will be explored theoretically during the coming contract period.

On the other hand, the largest droplet ($A = 0.254$ cm) does not seem to show the same steep dependence upon Reynold's number, and the values of the collection efficiency appear to cross over with those for smaller droplets. Although the larger droplet has hardly completely developed its motion and is still far removed from its terminal velocity, the crossover does seem strange. In order to resolve this, additional experiments are planned with an intermediate drop size ($A = 0.207$). Furthermore, data will also be obtained for an intermediate accelerating distance ($S = 165$ cm) in order to better delineate the transition between the curves for $S = 30$ cm and 298 cm in Fig. 8.

VI Extrapolated values

We stated at the outset that the goals of this research are to obtain the basic data necessary to calculate the rate of scavenging of atmospheric aerosol by rain as well as to explicate theoretically the mechanism of capture in fundamental physical terms. For the first of these goals we require collection efficiencies as a function of droplet size and particle size at the

appropriate terminal velocities assuming there are no additional effects due to electrical charge, particle shape, chemical composition, and atmospheric convection.

By extrapolation of the results in Fig. 9 for $A = 0.116$, 0.158 and 0.254 cm to the Reynold's numbers corresponding to the terminal velocities of these droplets ($Re = 1163$, 1933 , and 4063 , respectively), we obtain values of the efficiency for these size droplets of 5.0×10^{-3} , 2.1×10^{-3} , and 2.5×10^{-4} . These results are plotted in Fig. 11.

The magnitude of these collection efficiencies is much higher than those reported earlier for droplets which were still far removed from their terminal velocities. The present values lead to washout coefficients which will result in significant washout of atmospheric aerosol under real conditions. Additional values of the collection efficiencies at terminal velocity will be obtained for other droplet sizes and for various particle sizes from the additional experiments proposed earlier and designated on Fig. 2. These will then provide an adequate range of particle sizes and droplet sizes to enable us to calculate the washout coefficients of the submicron aerosol as a function of rainfall rates. The effect upon atmospheric visibility will also be calculated. These calculations will be carried out during the coming grant period.

References

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2. Zebel, G., J. Colloid Interface Sci. 27, 294 (1968).
3. Hampl, V., Kerker, M., Cooke, D.D., and Matijević, E., J. Atmos. Sci. 28, 1211 (1971).
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Table I

Entrance and exit velocities, Reynold's numbers and percentage of terminal velocity

Droplet radius cm	Acceleration Tube I						Acceleration Tube II						Acceleration Tube III						Terminal velocity	
	Entrance 30 cm			Exit 165 cm			Entrance 298 cm			Exit 433 cm			Entrance 515 cm			Exit 650 cm				
	cm/sec	Re	%	cm/sec	Re	%	cm/sec	Re	%	cm/sec	Re	%	cm/sec	Re	%	cm/sec	Re	%	cm/sec	Re
0.116	192.3	295	25.4	413.0	535	54.0	517.6	794	68.0	582.8	894	76.5	612.2	939	80.5	648.0	994	85.5	758	1163
0.158	193.0	405	21.0	429.3	897	46.5	548.5	1146	58.0	629.8	1316	68.0	667.7	1395	72.0	717.5	1500	77.5	925	1933
0.254	195.9	658	16.2	444.2	1492	36.8	579.7	1947	48.0	678.8	2280	56.0	727.0	2445	60.5	795.8	2673	66.0	1209	4063
0.116*	188.2	38	11.3	435.9	88	26.2													335	1662

*For helium gas; other values for nitrogen

Table II

Effect of particle size upon collection efficiency

droplet radius $A = 0.254$ cm
 acceleration tube 515 cm
 Reynold's number at entrance 2445
 Reynold's number at exit 2673
 Nitrogen

Sample	a μm	$E \times 10^5$	$Pe \times 10^{-8}$		Average	$I = (a/A) \times 10^4$
			Entrance	Exit		
442	0.348	20.8	8.51	9.31	8.91	1.37
446	0.353	21.8	8.66	9.47	9.06	1.39
449	0.360	21.6	8.86	9.69	9.27	1.42
451	0.340	18.8	8.28	9.05	8.66	1.34
452	0.340	20.2	8.28	9.05	8.66	1.34
455	0.136	36.5	2.54	2.78	2.66	0.55
456	0.135	47.5	2.51	2.75	2.63	0.54
458	0.148	25.0	2.86	3.12	2.99	0.58
459	0.180	19.6	3.73	4.08	3.90	0.71
461	0.167	24.8	3.37	3.69	3.53	0.66
462	0.161	29.5	3.21	3.51	3.36	0.64
464	0.229	19.9	5.09	5.57	5.33	0.90
468	0.241	20.7	5.43	5.94	5.68	0.95
469	0.241	20.1	5.43	5.94	5.68	0.95
474	0.133	40.8	2.46	2.69	2.57	0.52
477	0.117	40.4	2.05	2.24	2.14	0.46

Table III

Effect of particle size upon collection efficiency

droplet radius $A = 0.158$ cm
 acceleration tube 30 cm
 Reynold's number at entrance 405
 Reynold's number at exit 897
 Nitrogen

Sample	a μm	$E \times 10^5$	$Pe \times 10^{-8}$		Average	$I = (a/A) \times 10^4$
			Entrance	Exit		
398	0.284	2.22	1.10	2.44	1.77	1.80
402	0.296	2.60	1.16	2.57	1.86	1.87
403	0.258	5.30	0.98	2.17	1.57	1.64
404	0.294	5.90	1.15	2.55	1.85	1.86
407	0.195	16.90	0.68	1.52	1.60	1.23
411	0.166	9.30	0.55	1.23	0.89	1.05
412	0.160	16.70	0.53	1.16	0.84	1.01
413	0.171	16.10	0.57	1.28	0.92	1.08
414	0.211	10.01	0.76	1.68	1.22	1.34
416	0.378	2.23	1.55	3.44	2.49	2.39
417	0.436	2.15	1.83	4.07	2.95	2.76
418	0.464	3.01	1.97	4.37	3.17	2.94

Table IV

Effect of acceleration distance on collection efficiency

Droplet radius - 0.116 cm

Sample	$Ex \times 10^5$	$Pe \times 10^{-8}$		Aerosol modal radius μm	$I = (a/A) \times 10^4$	Acceleration distance cm
		Entrance	Exit			
141	1.2	0.60	1.37	0.316	2.724	30*
143	0.7	0.73	1.66	0.359	3.094	Re(entrance) 38
145	1.1	0.87	2.00	0.380	3.276	Re(exit) 88
146	0.8	0.87	2.00	0.380	3.276	
578	46.4	2.44	2.75	0.314	2.707	298
579	31.8	2.76	3.11	0.348	3.000	
580	36.0	2.76	3.11	0.348	3.000	Re(entrance) 794
581	54.7	2.76	3.11	0.348	3.000	Re(exit) 894
583	45.6	2.59	2.92	0.330	2.844	
586	50.8	2.79	3.14	0.351	3.026	
589	51.0	2.79	3.14	0.351	3.026	
590	196.0	2.71	2.87	0.298	2.569	515
591	313.0	2.71	2.87	0.298	2.569	Re(entrance) 939
593	174.0	2.79	2.95	0.305	2.629	Re(exit) 994

Table V

Effect of acceleration distance on collection efficiency

Droplet radius - 0.158 cm

Sample	Ex 10^5	Pe x 10^{-8}		Aerosol modal radius μm	I=(a/A)x 10^4	Acceleration distance cm
		Entrance	Exit			
398	2.22	1.10	2.44	0.284	1.797	30
402	2.60	1.16	2.57	0.296	1.873	Re(entrance) 405
404	5.90	1.15	2.55	0.294	1.861	Re(exit) 897
416	2.23	1.55	3.44	0.378	2.392	
542	30.3	4.04	4.64	0.352	2.227	298
541	20.7	4.04	4.64	0.352	2.227	
536	27.0	3.56	4.08	0.316	2.000	
533	45.2	3.99	4.58	0.348	2.202	Re(entrance) 1146
529	34.6	3.23	3.71	0.292	1.848	Re(exit) 1316
528	24.6	3.23	3.71	0.292	1.848	
527	21.2	3.53	4.05	0.314	1.987	
525	33.7	3.42	3.93	0.306	1.937	
511	96.7	4.31	4.63	0.315	1.994	515
509	76.4	5.04	5.41	0.359	2.272	
506	129.0	4.31	4.63	0.315	1.994	
501	45.8	5.14	5.52	0.365	2.310	Re(entrance) 1395
500	49.4	5.14	5.52	0.365	2.310	Re(exit) 1500
512	120.0	5.09	5.47	0.362	2.291	
513	36.4	5.09	5.47	0.362	2.291	
519	67.1	4.84	5.20	0.347	2.196	
521	68.8	5.07	5.45	0.361	2.285	

Table VI

Effect of acceleration distance on collection efficiency

Droplet radius - 0.254 cm

Sample	$Ex \times 10^5$	$Pe \times 10^{-8}$		Aerosol modal radius μm	$I = (a/A) \times 10^4$	Acceleration distance cm
355	14.5	Entrance	Exit	0.308	1.213	30
356	12.6	1.97	4.47	0.307	1.208	
357	14.7	1.97	4.47	0.307	1.208	
358	16.1	2.09	4.73	0.322	1.268	Re(entrance) 658
363	15.5	2.40	5.45	0.362	1.425	Re(exit) 1492
364	10.5	2.48	5.62	0.372	1.465	
365	16.5	2.04	4.65	0.317	1.248	
366	13.7	2.28	5.18	0.347	1.366	
367	11.8	2.28	5.18	0.347	1.366	
573	19.0	6.67	7.81	0.343	1.350	298
572	15.9	6.67	7.81	0.343	1.350	
571	16.3	6.04	7.07	0.316	1.244	Re(entrance) 1947
553	14.6	6.57	7.69	0.339	1.334	Re(exit) 2280
551	20.7	5.99	7.02	0.314	1.236	
547	20.6	5.99	7.02	0.314	1.236	
442	20.8	8.51	9.31	0.348	1.370	515
446	21.8	8.66	9.47	0.353	1.390	
449	21.6	8.86	9.69	0.360	1.417	Re(entrance) 2445
451	18.8	8.28	9.05	0.340	1.338	Re(exit) 2673
452	20.2	8.28	9.05	0.340	1.338	

Legends for Figures

Fig. 1 Schematic diagram of scavenging apparatus:

A, dropping device; B, accelerating tube, 30, 298 or 500 cm; C_1 , C_2 , photographic shutters; D_1 , D_2 , D_3 , sliding valves; E_1 , E_2 , valves; F_1 , F_2 , solenoids; G, timing circuit; H, 135 cm aerosol chamber; I, 37 cm lower chamber; J, funnel; K, filling tube.

Fig. 2 Acceleration height - droplet size domain. x -

experiments for average value of modal radius,

$a_M = 0.36 \mu\text{m}$. (x) - experiments for which modal radius

was varied. Δ - projected experiments with average value

of modal radius, $a_M = 0.36 \mu\text{m}$. $\odot$ - projected experiments for which modal radius will be varied.

Fig. 3 Collection efficiency versus particle radius for

$a = 0.254 \text{ cm}$, $S = 515 \text{ cm}$.

Fig. 4 Collection efficiency versus particle radius for

$a = 0.254 \text{ cm}$ and $S = 515 \text{ cm}$. Full line is least squares fit.

Fig. 5 Collection efficiency versus particle radius for

$a = 0.158 \text{ cm}$, $S = 30 \text{ cm}$. Δ - in nitrogen; $\circ$ - in helium

Fig. 6 Collection efficiency versus particle radius for

$a = 0.158 \text{ cm}$, $S = 30 \text{ cm}$. Full line is least squares fit.

Fig. 7 Collection efficiency versus Peclet number for $a = 0.254$ cm, $S = 515$ cm in nitrogen ($\circ$), $a = 0.254$ cm, $S = 30$ cm in helium (Δ), and $a = 0.158$, $S = 30$ cm in nitrogen ($\square$). The full lines are least squares fits to the data. The dashed lines are Suneja's theory for $I = 0$ and 10^{-4} . The dotted line is Zebel's theory for creeping flow.

Fig. 8 Collection efficiency versus particle radius for accelerating distances $S = 30, 298, \text{ and } 515$ cm. Average value of $a_M = 0.36 \mu\text{m}$.

Fig. 9 Collection efficiency versus Reynold's number for droplet radii $A = 0.116, 0.158, \text{ and } 0.254$ cm. Average value of $a_M = 0.36 \mu\text{m}$.

Fig. 10 Collection efficiency versus Péclet number for droplet radii $A = 0.116, 0.158, \text{ and } 0.254$ cm. Average value of $a_M = 0.36 \mu\text{m}$.

Fig. 11 Value of collection efficiency extrapolated to terminal velocity for $A = 0.116, 0.115 \text{ and } 0.254$ cm.

FIG 1

FIG 2

FIG 3

FIG 4

FIG 5

FIG 6

FIG 7

File 8

FIG 9

Fig 10

FIG 11

A