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A.

MEASURIMENT OF THE RETENTION OF VARIOUS AEROSOLS IN THE RESPIRATORY TRACT USING A SCATTERED-LIGHT PARTICLE-COUNTER

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I. a) Introduction

Assessment of the biologically dangerous impurities in the atmosphere is not directed solely at the chemical peculiarity of the materials in suspension. To the contrary, physical characteristics and properties are also of decisive significance. In this connection, exact information about the depth of penetration and the deposition of aerosols in the respiratory system is of fundamental importance for aerobiological problems. This stands to reason all the more if the different construction of the various mucosa components in the respiratory system as well as their special function is taken into consideration. How many particles of a certain fraction from the aerosol spectrum are laid down on specially resorptive membranes is therefore decisive as regards the biological hazard of an aerosol that is injurious to the health. The ability of an aerosol to penetrate the airways and the deposition of the particles depend essentially on the size of the individual particles. FINDEISIN calculated purely theoretically what size aerosol particles must have in order to be deposited in rather large number if certain sections of the lungs. In approximation of anatomical conditions he tock into consideration Brownian molecular movement, the force of gravity and flow conditions in the bronchi. He found that the optimum particle size for an aerosol's penetration of the airways was a diameter between 0.1 and $5\,\mu$. Further factors such as temperature and moisture in the air passages as well as hygroscopic behavior, surface properties and electrical charge of the particles can alter sedimentation conditions.

b) The Scattered-light Method of Measurement

Aercsol research has for many years been concerned with knowledge about the retention of colloidal systems, especially in the respiratory tract. Research results have in part been very different so that a reexamination using the scatteredlight method of measurement seemed purposeful. During our experiments the retention of a few, physically and chemically very different aerosols in the respiratory tract was measured. The most important device in these measurements was a scatteredlight particle counter (ROYCO), which made it possible to evaluate aerosol spectra in a few minutes. The registration of particles and the simultaneous measurement of particle size take place in the following manner (Figure 1):



Prinzip des Streulicht-Messverfahrens

Figure 1. Working principle of scattered-light method of measurement

Xeys:

- 1. Lens system
- 2. Lamp
- 3. Measuring element

A small amount of an aerosol that is to be investigated is sucked away and conducted through a measuring element. In this element the aerosol flow crosses an intensive light beam. The passing aerosol particles scatter the light at a right angle to the direction of the light beam into a photomultiplier which transform the flashes of light into electrical pulses. The intensity of the flash of light that is produced when an individual particle crosses the light beam is a function of particle size, so that the pulses of the multiplier characterize particle size. The electrical pulses are sorted according to size, are counted and the result printed cut. Depending on the density of the aerosol, an aerosol spectrum ranging from 0.1 to 5 μ can be run through in 0.3 to 10 minutes of measuring time.

c) Equipment for Measurement of Retention

The equipment for measuring the retention of aerosols in the respiratory system consists essentially of three parts (Figure 2):

- 1) atomizer or aerosol generator with dilution system and aerosol channel;
- 2) mouthpiece with one each electromagnetically controlled inspiration and expiration valve;
- 3) expiration or measurement channel with particle counter.

Serving as aerosol generator is a HEYER ring nozzle, operated by compressed air. The density of the aerosol in the aerosol channel can be varied by means of a dilution system with the addition of filtered compressed air in the ratio of 1:100. Atomizer and dilution system are so proportioned that 50 liters of aerosol per minute are always delivered into the aerosol channel. Maximum inspiration requirement can be provided from reserve pipe so that respiration mechanics are not impeded even in case of hyperventilation. The concentration and the aerosol spectrum can be kept practically constant for several minutes.

The electromagnetic inspiration- and expiration-valves on the mouthpiece are controlled by a step-by-step relay. An indicator lamp, operating synchronously with the step-by-step relay, informs the experimental person of the beginning of the inspiration and expiration phase. The step-by-step relay is powered by a pulse generator of fixed-adjustable frequency. Thus, on the one hand, the subject is obliged to breathe uniformly while, on the other hand, it is easily possible to open the expiration valve within a certain interval of the expiration phase only and to fill the measuring pipe with re-

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spired air of precisely this fraction of the expiration phase. The remaining expired air escapes into the atmosphere (if the inspiration - and expiration valve is closed).



Figure 2. Experimental set-up for measurement of retention

L.	At Cm126r	9.	Measuring channel
2	Dilution system	10	Warm air blower
3	Acrosol channel	11	Warm air
4	Reserve pipe	12	Scattered-light parti-
Б.	Monthpiece	•	cle counter
6	Safety valve	13.	Step-by-step relay
7.	Inspiration valve	14.	Current supply for
8	Expiration valve		solenoid

The measuring channel, to which the particle counter is connected, is kept at a temperature of about 38° C in order to avoid condensation of the respired air. In the measuring channel there is a small, slowly operating ventilator which assures sufficient intermixture of the acrosol in the channel. The volume of the measuring channel is about four liters.

II. a) Spectra and Total Retention of Latex Aerosols

Retention of later aerosols was investigated first, Suspensions of spherical later particles of exactly defined particle size are available on the market and can serve for

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the production of monodispersed aerosols. The scattered-light particle counter was calibrated with such monodispersed latex aerosols. Naturally, the measurement error of the scatteredlight particle counter is less in the case of monodispersed aerosols consisting of spherical particles than in the case of aerosols of a different kind. For this reason and because latex aerosols of low concentration are harmless besides, retention measurements were conducted first with latex aerosols.

The procedure for retention measurement was as follows. First of all, the aerosol spectrum produced by the atomizer was measured. For this purpose the inspiration- and expiration-valve was opened, and the reserve pipe and mouthpiece closed. The aerosol flowed from the atomizer through the aerosol channel and the mouthpiece into the measuring pipe. After about a minute both valves were closed and the atomizer turned off. The particle counter sucked away a part of the aerosol in the measuring pipe and registered the particle spectrum.

During measurement the pulses delivered by the photomultiplier of the particle counter were observed oscillographically. In order to avoid incorrect measurements, the density of the aerosol was so adjusted that the multiplier pulses did not follow one another too closely. In all the experiments the particle density of the aerosol was about 100 particles per cubic centimeter.

After measurement of the spectrum, all the equipment was rinsed with filtered compressed air. Immediately afterwards the atomizer was set in motion again, and the aerosol flowing through the aerosol channel inhaled through the mouthpiece and exhaled into the measuring channel. The valves were automatically opened and closed, as just described. Respiretory rhythm was thus fixed. In all the measurements inspiretion and expiration took place by the mouth. Breathing through the nose was precluded by a nose clamp. In each instance 20 breaths were taken per measurement. Respiration frequency was always 17 breaths per minute. On termination of the experiment the expired aerosol in the measuring pipe was measured.

Figure 3 shows a particle spectrum of a latex aerosol whose particles (according to information from the producer) have a diameter of 1.305μ . Curve I shows the aerosol spectrum produced by the atomizer, Curve II the spectrum of an expired 1.305μ latex aerosol. Particle diameter in μ is plotted on the abscissa, and particle number per cubic centimeter of air on the ordinate. The abscissa is graduated linearly, the ordinate logarithmically.

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Figure 3. Later aerosol spectra

Xoys:

 Curve I: Aerosol spectrum of a 1,305 μ latex aerosol
 Curve II: Spectrum of an expired latex aerosol

3. Particle 0.1 \u03c4 channel width 30 cm³

4. Particle diameter à

Figure 4 shows acrosol spectra of a 0.557 /4 latex acrosol.

From measuring the aerosol inspired and expired, the amount of the aerosol deposited in the respiratory system or the retention can be calculated. (If the particle counter in measuring the aerosol spectrum produced by the atomizer in a definite size interval, e.g. in the interval 0.5-0.6 μ , had counted N₁ particles, and N₂ particles during measurement of the expired aerosol in the same size interval, the percentage retention of aerosol particles of this size

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R . X1 - 37 . 100)





Keys:

- 1. Curve I: Spectrum of a 0.557 / latex aeroch 2. Curve II: Spectrum of an expired 0.557 / latex
- aero so 1
- 3. Particle
- 0.1 µ channel width 30 cu cm -
- 4. Particle diameter d

Figure 5 shows the retention of latex acrosols as a function of particle size. Indicated on the abscissa is particle diameter in μ , and on the ordinate retention in percentages. It can be seen that retention for latex particles between 0.2 and 0.4 μ diameter is about 30%. Retention increases constantly with particle diameter. For particles between 3 and 4 μ it is about 90%. The plotted measures values are mean values from five measurements in each instance. Conditions of the experiment were: inspiration and expiration through the mouth; 17 deep breaths per minute.

The curve agrees very well with a retention curve measured by VIJK and PATTERSON (FUCHS, The Liechanics of Aurosols, page 236).





Key: 1. Particle diameter d

All that can be inferred from this retention curve is what percentage rate of particles of a certain size was deposited in the respiratory system altogether. As for the site of the deposition, no statement can as yet be made from this measurement. In further experiments, therefore, localization of the acrosols deposited in the bronchial system would have to be clarified.

b) Retention of Different Practions of Respired Air

In the course of an expiration the respired air increasingly contains fractions from deeper sections of the lungs.

The respired air originating from the bronchial system equations in the first fractions fractions originating from the large bronchi, whereas at the end of the expiration alveolar air is exhaled. Now with our experimental set-up it is possible to measure the aerosol spectra of different fractions of expired air. The deposition site of the aerosol can be judged approximately from the differing aerosol concentration of successive fractions of expired air.

For a rough determination of the aerosol deposition site

in the respiratory system the explication phase was divided into four large intervals of equal time, or the expired air was divided into four fractions.

To begin with, retention of the air exhaled in the first quarter of the expiration phase was measured. In the process the expiration value of the step-by-step relay was controlled so that it opened only during the first quarter of the expiration phase. Only the air expired during this time reached the measuring channel; the air expired afterwards flowed via a safety value from the mouthpiece into the atmosphere. After 20 uniform breaths the measuring channel was filled with air from the first quarter of the expiration phase, and the aerosol spectrum of this air could be measured. The procedure was the same with the other three intervals of the expiration phase.





Xeys:

- Measurement during different intervals of the expiration phase
 Expiration phase

Altogether three series of measurements were conducted, in each instance with a monodispersed latex serosol, viz.

1. with a 0.365 µ latex aerosol,

2. with an 0.871 μ and

3. with a 2,68 / latex aerosol.

Figure 6 shows the result of this measurement. The retention of 0.365 μ latex aerosol reveals a uniform increase in retention from quarter phase to quarter phase. This makes us conclude that there is uniform distribution of deposition over the entire respiratory system.



Figure 7. Differential retention of latex aerosol.

Key: 1. Expiration phase

The 0.871 / aerosol shows 50% greater retention in the first quarter phase than does the 0.365 / aerosol. The larger particles thus are already deposited more intensely in the upper respiratory tract. This is shown even more clearly in the case of 2.68 / latex aerosol. Already 87% of the inspired aerosol is deposited in the upper airways. The slight increase in retention of the following fractions of expired air shows that only relatively few of the 2.68 / particles were able to penetrate more deeply into the respiratory system. This fact can be gathered more easily from the measurement results if the increase in retention of successive fractions is plotted in a diagram. Figure 7 presents this retention increase, which might also be called "differential retention." "Differential retention" can be taken as a measure of the deposition in different perts of the respiratory system. When the three diagrams are compared, it can be perceived that retention of the first fraction rises sharply with increasing particle radius, whereas retention of the last fraction at first rises with increasing particle radius and then falls again. Thence it can be concluded 1) that the increase of total retention with increasing particle diameter is caused mainly by increasing deposition in the upper airways and 2) that deposition in the alveoli must have an optimum value for later particles with a diameter of about 0.9 μ . Later particles with a diameter larger than 3 μ in practice hardly get as far as the alveoli.

c) NaCl Aerosol Spectra

A hygroscopic ærosol in the respiratory tract behaves in a fundamentally different way from a latex aerosol. The particle size of a hygroscopic aerosol, say an aerosol made up of NaCl particles, is highly dependent on the moistness of the gaseous phase in which it happens to be. This can easily be observed with the scattered-light particle counter.





Keys:

- Curve I: NaCl aerosol in dry air. Dilution 1:20.
 Curve II: NaCl aerosol in 95% moist air (dilution 0).
- 3. Particle diameter d

If, for example, a 1-molar NaCl solution [See Note] is atomized and if by admixture of about 95% moist mixed air it is assured that the NaCl aerosol particles in the moist air of the atomizer arrive in the particle counter, an aerosol spectrum is measured, as shown by Curve II in Figure 8. The spectrum obviously has a maximum at 0.4 μ . It extends as far as 5μ .

[Note]: In the NaCl aerosol experiments a 1-molar NaCl solution was always a tomized.

The possibility existed of admixing filtered to the aerosol on the way from the measuring channel to the particle counter in order to reduce the aerosol concentration. Now if dry hot air is admixed to a moist NaCl aerosol in the ratio of 1:10 or 1:20, apart from the dilution of the aerosol a shift of the spectrum towards small particle diameters is observed, Curve I shows the spectrum of an aerosol diluted with dry hot mixed air in the ratio 1:20. The density of this aerosol in the measuring channel, however, by virtue of an increase in the amount of aerosol generated by the atomizer was greater by a factor of 20 than the density of the aerosol with the spectrum represented by Curve II. Comparison of both spectra, therefore, causes us to perceive only a shift of the spectrum towards small particle diameters. It is to be noted that the rise of Curve I is about six times as great as that of the straight part of Curve II. This means that the linear measurements of the NaCl aerosol particles are about six times greater in 95% moist air than in dry air. Enlargment of the NaCl aerosol particles in the moist respired air can likewise be Observed.

Figure 9 shows three aerosol spectra.

Curve I shows the spectrum of an NaCl aerosol which was in dry air from the atomizer to the particle counter and which was additionally diluted between the measuring channel and the particle counter with dry hot air in the ratio 1:10.

Curve II shows the spectrum of an expired NaCl aerosol diluted with dry hot air in the ratio 1:10. It may be assumed that during measurement of both these spectra the NaCl aerosol particles flew through the measuring chamber of the particle counter as practically dry salt crystals, i. e. NaCl crystal aerosols were observed.

Curve III shows the spectrum of an expired NaCl aerosol without the addition of dry mixed air, i.e. the measurement was made in moist respired air. This spectrum reveals a maxi-

mum at 0.6 μ and extends as far as 5 μ . Comparison of spectra II and III shows that the NaCl particles are about six times larger in moist respired air than in the dried state.

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Keys:

- Curve I: Spectrum of an NaCl aerosol measured 1. in dry air (dilution 1:10)
- Curve II: Spectrum of an expired NaCl aerosol in 2. dried air (dilution 1:10) Curve III: Expired NaCl aerosol measured in moist
- 3 air (dilution 0)
- 4. Particle diameter d

d) Retention Measurements with NaCl Aerosols

The procedure during measurement of total retention of NaCl aerosols in the respiratory system was, fundamentally, exactly the same as during the corresponding latex aerosol measurements. The NaCl aerosol was in dry air from the atomizer up to the mouthpiece. Thus in practice NaCl crystals were inhaled. Curve I shows the spectrum of such an NaCl aerosol. The diameters of these particles grow about six-fold in the moist respired air, as comparison of spectra II and III showed. In order to determine what fraction of the inspired

NaCl crystals was deposited in the respiratory system, the expired moist NaCl acrosol particles have to be put back in the state they were in before inspiration. This is done, as already described, by admixing hot dry air to to the expired NaCl acrosol en route between the measuring channel and the particle counter.



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Key: 1. Particle diameter d

Figure 10 shows the result of such a retention measurement. The plotted measured values are mean values, usually from five retention measurements. The form of the curve is similar to the retention curve of latex aerosols.

Of interest is a comparison of the NaCl retention curve with the latex retention curve. Figure 11 presents a comparison of these curves. If the particle-size values of the NaCl retention curve are multiplied by the factor 6, a curve is obtained which practically coincides with the latex retention curve. By reason of their growth in the moist air of the respiratory passages the inspired NaCl crystals in the respiratory system thus behave in the same way as do latex particles six times as large. DAUTREBAND and WALKENHORST (C. N. DAVJES, Inhalad Particles and Vapours, page 117) come to similar experimental results.





Keys:

1. Transformed NaCl curve 2. Particle diameter d

Then, just as in the case of the latex aerosol, NaCl retention of different phases of expired air was measured. Figure 12 shows the retention of four fractions of the expired air for NaCl crystals with a diameter of $0.15-0.2\,\mu$ (Curve I) and for NaCl crystals with a diameter of $0.3-0.4\,\mu$ (Curve II). For comparison, the corresponding curves for an $0.871\,\mu$ latex aerosol and a 2.68 μ latex aerosol are drawn in (Curves III and IV). The NaCl retention curves lie between the latex curves. This is understandable if it is assumed that the NaCl aerosol particles in the respiratory system behave as do latex particles about six times as large. A 1 μ latex aerosol would then correspond to the $0.15-0.2\,\mu$ NaCl aerosol.

From the relatively high retention of the first fraction the conclusion can be drawn that NaCl particles grow already in the upper regions of the respiratory system so that a great part of them is already deposited there.

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Figure 12. Retention of NaCl aerosol in the respiratory tract, measured in different intervals of the expiration phase.

Keys:

 Curve I: Particle diameter: 0.15 - 0.2 μ Curve II: Particle diameter: 0.3 - 0.4 μ For comparison Curve III: Latex 0.871 μ Curve IV: Latex 2.68 μ

2. Expiration phase

e) Measurement of the Retention of Cigarette Smoke

It was also attempted to measure the retention of cigarette moke. The difficulty in the measurements is that the acrosol spectrum of a nigarette changes so rapidly that reproducible measurements are possible only at great cost. Measurements of the retention of cigarette smoke are full of relatively large errors.

Figure 13 shows first two spectra of cigarette smoke. Curve I shows the acrosol spectrum of a cigarette without a filter, and Curve II the spectrum of a filter cigarette. The acrosol particles are smaller than $1 \ \mu$, with the maximum of the spectrum probably lying in the vicinity of $0.1 \ \mu$. Fig-

ure 14 shows the measured retention values for cignrette smoke. They are in the vicinity of the latex retention curve.



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Figure 13. Aerosol spectra of cigarette smoke.

Keys:

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1. Curve I: Cigarette without filter

- 2. Curve II: Cifarette with filter
- 3. Particle
 - 0.1 1 channel width 30 ou om
- 4. Particle diameter d

On the basis of these examples we can show that an urgent problem in acrosol research, viz, knowledge as to the deposition of particles of differing schavior in the respiratory tract, can be clarified by means of our experimental procedure within certain limits of error. Sithin the framework of such investigations, however, the dynamics of the transformation of acrosol systems in the respiratory tract also deserve the special attention of research. Meanwhile, we have succeeded in continuously encompassing the momentary reaction phase, measured on the particle-size spectrum. By this means, possibly, in future interesting perspectives on the transformation processes of aerosol systems in the respiratory tract will be obtained, and therewith assessment of the hazard of an aerosol -- at least as far as its physical quality is concerned -- is possible.

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Koys:

1. Curve I: Retention of cigarette amoke 2. Curve II: Retention of latex aerosol 3. Particle diameter d

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