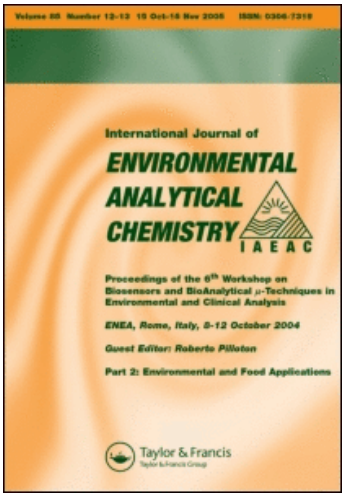

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Development of a Preseparator for Denuder Sampling

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Development of a Preseparator for Denuder Sampling

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Dedicated to Prof. W. Haerdi on the occasion of his 60th birthday.

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A simple aerosol preseparator was developed and its size classifying characteristics evaluated. The preseparator operates as a combination of a virtual and a real impactor. Two final versions are presented, both possessing an effective cut-off diameter (ECD) of about 2.3 μm. They are designed for combination with denuder sampling.

INTRODUCTION

For almost 30 years so called denuder tubes have been used successfully for diffusional separation of gaseous and particulate matter in air sampling and analysis (Pack *et al.*, 1959; Weinstein and Mandl, 1971; Bailey *et al.*, 1976; Gregory and Moyer, 1977; Ferm, 1979; Niessner and Klockow, 1980; Slanina *et al.*, 1981; Niessner and Klockow, 1982). Compared to sampling with filters, denuder tubes have the advantage of minimizing interactions between reactive compounds during the enrichment step. This technique is now

frequently used to collect ammonia and acidic airborne material, whereas filter sampling of these species is strongly subjected to artifact formation (Klockow *et al.*, 1979).

It is a well known fact that ammonium salts and particulate acidic constituents of the atmospheric aerosol predominantly occur in the so called accumulation mode (Junge and Scheich, 1969; Brosset *et al.*, 1975; Marlow and Tanner, 1976; Tanner *et al.*, 1979), with particle diameters below 2.5 μm (respirable fraction). It can be shown (Ferm, 1979; Niessner, 1981) that particles of this size range are not deposited in properly designed and operated denuder tubes whereas gases are almost completely retained if suitable wall coatings are used. Larger particles, however, may be lost by gravitational settling and consequently contribute to the results obtained in the analysis of denuder tube wall coatings. To avoid such an interference the tubes should be employed in a vertical position only. An additional remedy is the use of a size classifying preseparator which allows only particles below about 2.5 μm aerodynamic diameter to enter a denuder tube (Lippmann, 1976). By this means also turbulent losses of larger particles at the entrance of a tube are minimized.

Accordingly the preseparator to be developed should exhibit the following characteristics:

1. An effective cut diameter (ECD) between 2–3 μm with a sharp particle fractionation.
2. No real impaction plate because of the possibility of topochemical reactions between the impacted particles.
3. A design such that the connection from the aerosol inlet to the entry of a denuder tube is as short as possible, thus minimizing diffusion losses of gaseous acids in the preseparator.

These requirements are only fulfillable by the "opposing jet"-method (Willeke and Pavlik, 1980) and the "virtual" impactor principle (Loo *et al.*, 1976; Dzubay *et al.*, 1977; McFarland *et al.*, 1978). Particle-size classification by the opposing jet method needs a rather complicated design and the combination with a denuder tube is difficult to be accomplished. At the other hand, the combination of a virtual impactor with the inlet of a denuder tube is easy to be achieved. A detailed theoretical description of the behaviour of a virtual impactor is given by Marple and Chien (1980).

It was the purpose of this work to develop a simple single stage virtual preseparator for use in combination with denuder tubes and to evaluate its collection and separation characteristics as a function of operational and design parameters.

EXPERIMENTAL SECTION AND RESULTS

From a first version (Nießner, 1981) and a further development (Plomp, 1981) two final versions (Preseparator I and Preseparator II) were designed.

Preseparator I

The constructional details of Preseparator I (stainless steel) are given in Figure 1. Coming from the aerosol inlet with accelerated air stream Q_1 is divided into two flows, one with a minor flow rate Q_2 and the second with the major flow rate Q_3. The particles are injected into the coarse particle outlet Q_2, which has a larger diameter thus forming a flow stagnation zone. Only the larger particles are able to penetrate this zone due to their inertia. Smaller particles are drawn into the fine particle outlet Q_3 at a flow ratio of Q_3/Q_1. Between the axes of acceleration inlet nozzle and the fine particle outlet there is an angle of 60°. For the proposed low-volume sampling the total flow rate $Q_1 = Q_2 + Q_3$ was chosen to be in a range between 210 l/h and 400 l/h with a ratio of $Q_3/Q_2 = 10$.

As a test, artificial sea-spray aerosol created by the bursting bubble mechanism was applied. The generation system was described by Mallant *et al.* (1978). The sea-salt test aerosol is especially suitable because of the intended use of the preseparator/denuder-combination in coastal areas. The aerosol resulting from the "micro-ocean" typically has a bimodal size distribution.

Calibration of the preseparator was accomplished by use of a Royco 225 optical particle counter. A schematic diagram of the aerosol generation and calibration arrangement is shown in Figure 2. The bubbles are generated by blowing clean air through an immersed fritted glass filter at an overpressure of 0.35 bar. The aerosol is mixed with 1.2 m^3/h of dried air. From this aerosol stream a definite portion was used to determine the preseparator cut-off

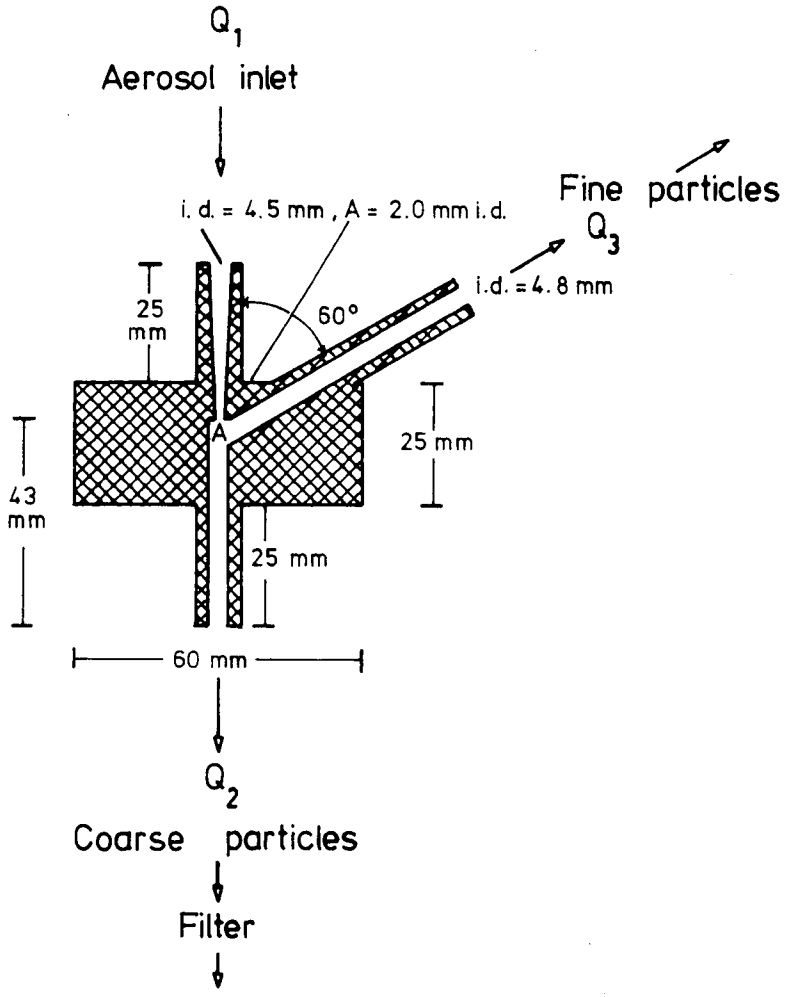

Figure 1 Schematic view of Preseparator I.

characteristic by comparison of the particle number distribution with and without the preseparator. The Royco 225 always measured the size distribution of particles in the major flow, Q_3, because this aerosol flow is the important one for denuder sampling. The whole set of results is graphically presented in Figure 3. Given is the fractional penetration for a certain size range (in flow Q_3) as a function of flow rate Q_2 and flow rate Q_3. The ECD is around 2.3 μm diameter.

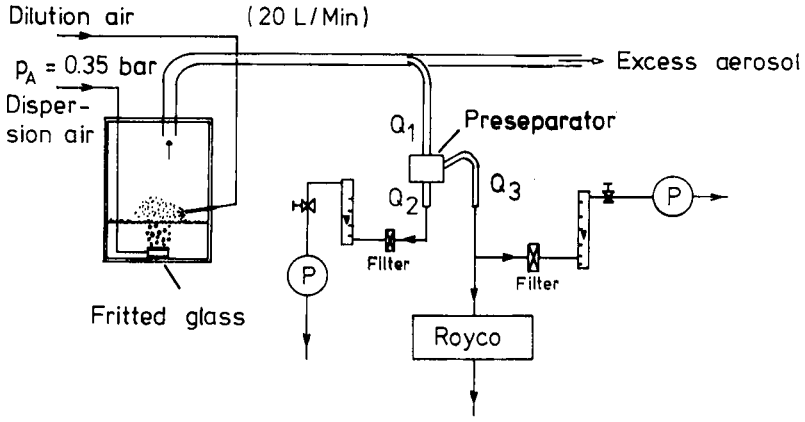

Figure 2 Experimental set up for production of artificial sea spray aerosol and calibration of the preseparator.

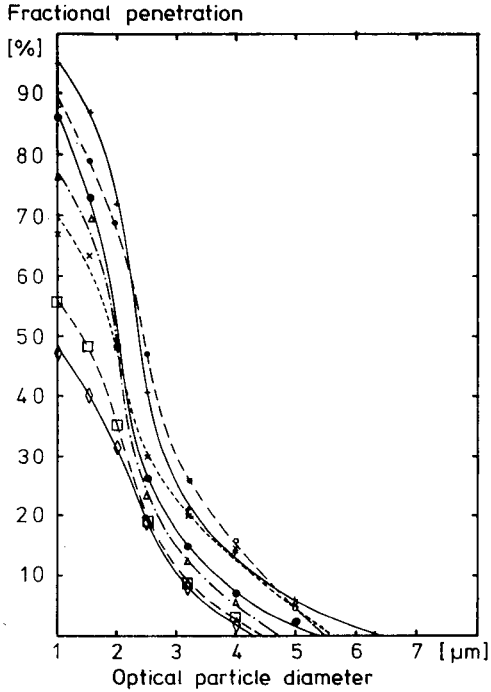

Figure 3 Fractional penetration of particles through Preseparator I (in flow Q_3) as a function of optical diameter at various flow rates Q_1 and flow ratios Q_2/Q_3: ◇ $Q_1 = 14$ L/min; $Q_2/Q_3 = 1/13$, □ $Q_1 = 11$ L/min; $Q_2/Q_3 = 1/10$, × $Q_1 = 7$ L/min; $Q_2/Q_3 = 1/6$, △ $Q_1 = 6$ L/min; $Q_2/Q_3 = 1/6$, ● $Q_1 = 5.5$ L/min; $Q_2/Q_3 = 0.5/5$, ○ $Q_1 = 4.5$ L/min; $Q_2/Q_3 = 0.5/4$, + $Q_1 = 3.5$ L/min; $Q_2/Q_3 = 0.5/3$.

An identical preseparator made of Akulon® was independently calibrated with monodispersed DOP-aerosol produced by the spinning disk aerosol generator (Model 8330, Research Appliance Company; Gibsonia/Pa., USA). The DOP aerosol was labelled with 0.5% Uranine for fluorimetric analysis (Rao, 1975). The aerosol of each flow, Q_2 or Q_3, was collected by means of Teflon® filters (LSWP, Millipore Corp.). These filters were selected because of their low blank values in the fluorimetric analysis. The extraction of filter and preseparator was accomplished by treating them in a 1:1 mixture of isopropanol and bidest. water in an ultrasonic bath for 15 min. The penetration of particles through the impactor as a function of aerodynamic particle size is given in Figure 4. In this case the ECD is around 1.7 μm diameter. From the detailed values of DOP deposition, given in Table 1, one can see that in case of an aerosol with a high sticking probability, like DOP, larger particles are preferably impacted on the walls of the duct of flow Q_2. This indicates that the preseparator operates not only on the basis of virtual impaction but also on real impaction of the larger particles. Compared to a real impactor there exists, however, one advantage: The impaction enrichment takes place mainly in the conducting tube of Q_2 and possible secondary reaction products are pumped off with

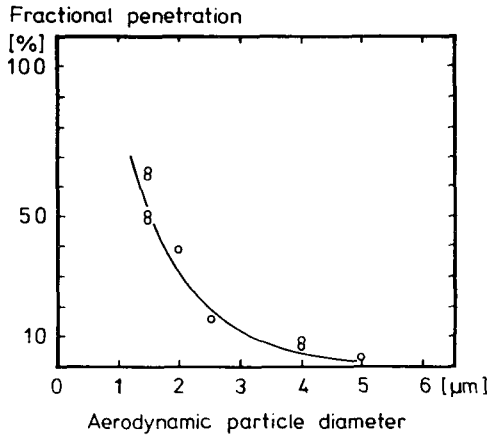

Figure 4 Fractional penetration of monodisperse DOP-particles through Preseparator I (in flow Q_3) as a function of particle diameter. $Q_1 = 5.0$ L/min; $Q_2/Q_3 = 0.5/4.5$.

Table 1 Average aerosol mass distribution in Preseparator I as a function of particle aerodynamic diameter. $Q_1 = 5.0$ L/min, $Q_2/Q_3 = 0.5/4.5$

Aerodynamic particle size	1.5 μm		2 μm		2.6 μm		4 μm		5 μm	
	$\bar{M}$ (%)	*SD* (%)	$\bar{M}$ (%)	*SD* (%)	$\bar{M}$ (%)	*SD* (%)	$\bar{M}$ (%)	*SD* (%)	$\bar{M}$ (%)	*SD* (%) (1)
Fine particle flow Q_3	57.2	14.8	38.9	5	15.3	9.7	8.1	13.1	3.5	—
Coarse particle flow Q_2	4.5	26	4.3	19	1.5	110	3.5	6.1	5.2	—
Preseparator walls	38	23	56.9	1.9	83.3	3.8	88	0.6	91.4	—
Sum of aerosol mass of Q_2 plus preseparator walls	42.5	20.4	61.2	0.2	84.8	1.8	91.5	0.4	96.4	—

$\bar{M}$ = Average aerosol mass.
SD = Standard deviation.
(1) one experiment.

Q_2 such having no influence on the composition of the fine particle flow Q_3.

Based on these results a final version of preseparator I, made of Kel-F® (because of its minimal adsorption of gaseous HCl and HNO_3) was built and combined with a Teflon® wind shield. The coupling to the denuder tube was accomplished by an isokinetic flow divider (see Figure 5).

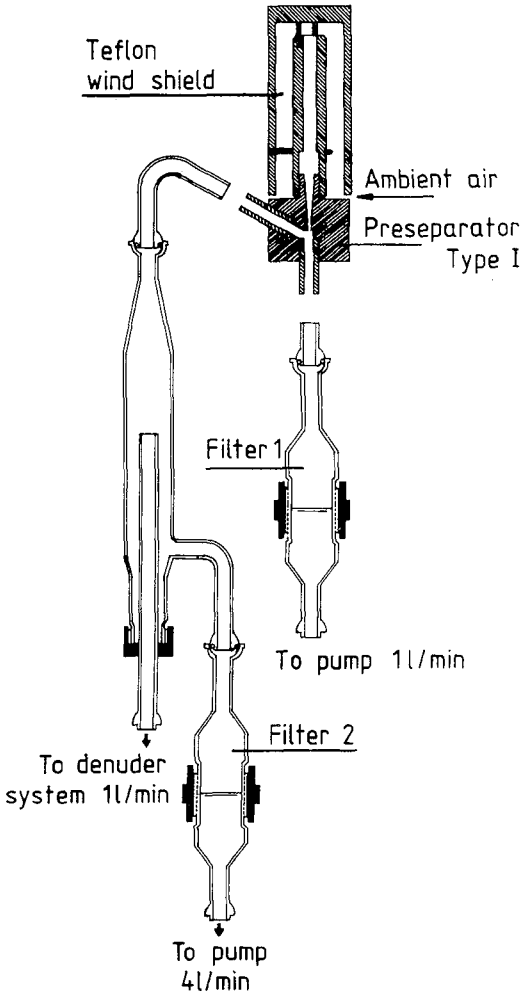

Figure 5 Experimental arrangement of the wind shielded Preseparator I connected to a denuder system for field measurements.

Preseparator II

For combination with a linearly arranged multidenuder system (Niessner and Klockow, 1982) a preseparator with an ECD of about 2.5 μm at a flow rate $Q_3 = 120$ l/h was required. The first denuder tube had to be inserted directly into the preseparator housing in order to minimize unwanted diffusion losses of gaseous HCl and HNO_3 prior to entering the denuder system. From the results obtained during the development of preseparator I an inlet nozzle with a diameter of 1.5 mm was selected to achieve the required ECD. The flow velocity within this nozzle is 18.9 m/sec.

The constructional details are given in Figure 6. As can be seen, the connection to the denuder tube (9 mm outer diam.; 6 mm inner diam.) is accomplished by a construction consisting of a screw cap and a short piece of silicon tubing as an O-ring, which seals the glass tube. All inner walls which come into contact with the aerosol are made of low porosity Teflon®. To improve its mechanical stability the preseparator is completely covered by a stainless steel housing.

Preseparator II was tested with a dry polydisperse Fe_2O_3-aerosol. The experimental arrangement is presented in Figure 7. The Fe_2O_3 powder was dispersed by means of a fluidized bed aerosol generator

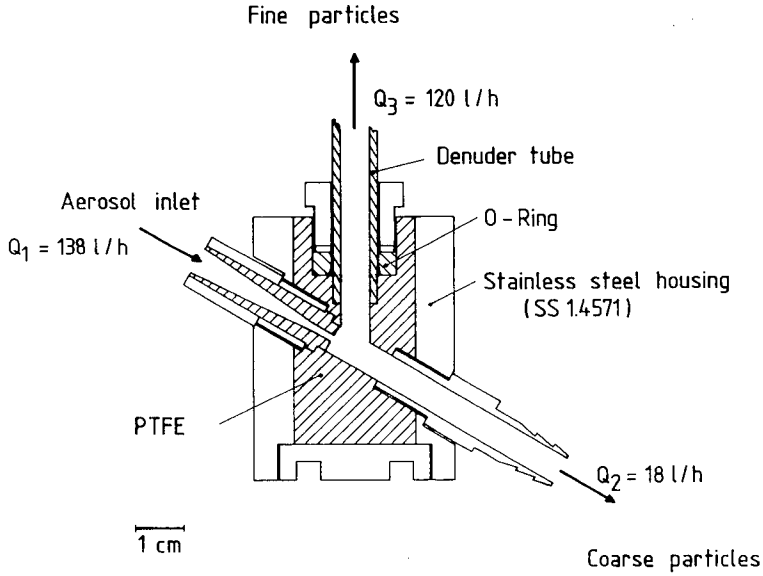

Figure 6 Schematic view of Preseparator II.

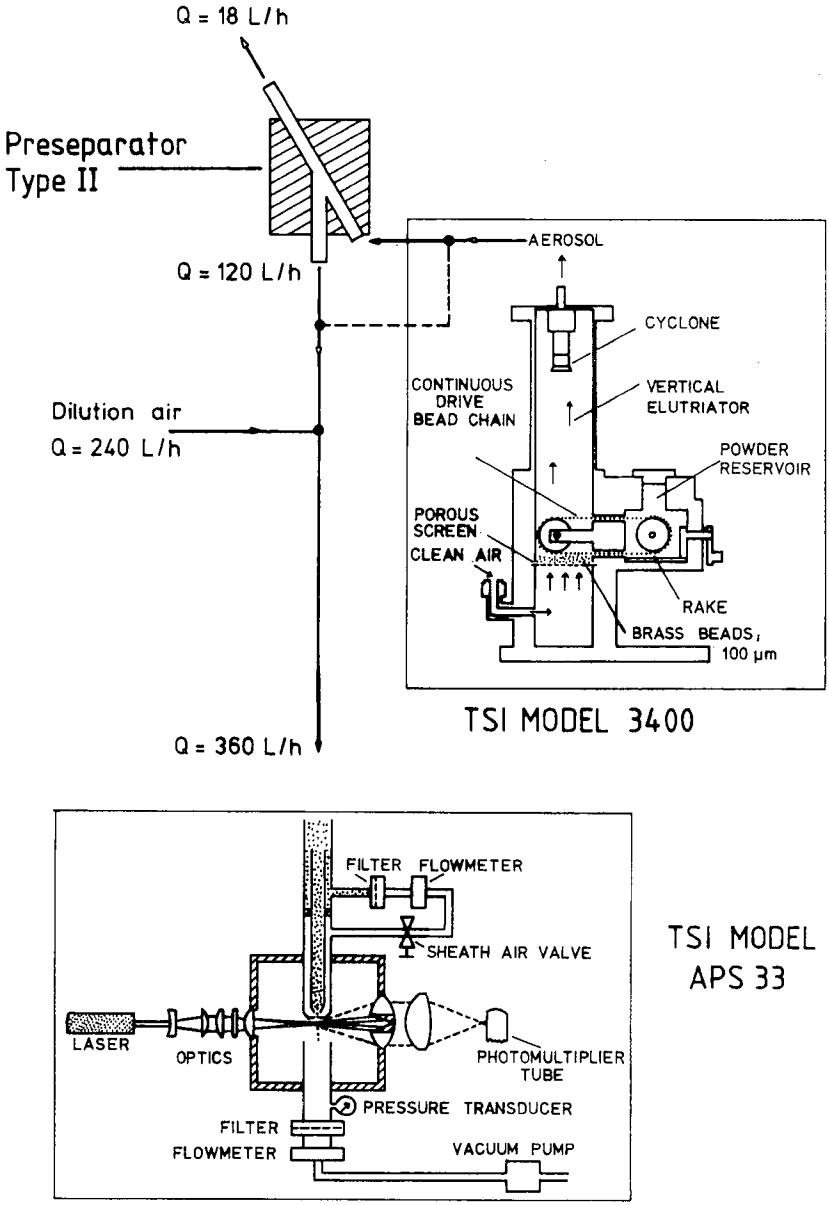

Figure 7 Experimental set up used for calibration of Preseparator II by use of a polydisperse Fe_2O_3 aerosol and an APS 33 optical particle detector.

(TSI Model 3400). From the aerosol generator outlet the aerosol was drawn directly through the preseparator II into the Aerodynamic Particle Sizer (TSI Model APS 33). Within 50 sec measuring time the whole size distribution was obtained by computer assisted data evaluation. Comparing the number size distributions up- and downstream of the preseparator allows a quick determination of the size fractionation characteristics. The result—as fractional penetration with flow Q_3—is plotted in Figure 8 and confirms the ECD predicted from the experiences gained with preseparator I.

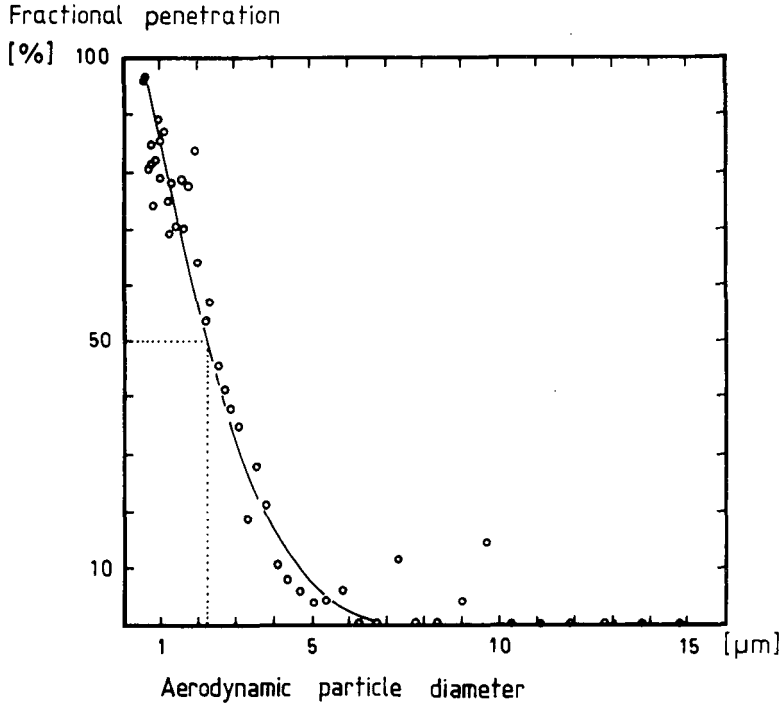

Figure 8 Fractional penetration of Fe_2O_3 particles through Preseparator II as a function of particle diameter. $Q_1 = 138$ L/h; $Q_2/Q_3 = 18/120$.

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References

Bailey, R., Field, P. and Wightman, G. *Anal. Chem.* **48,** 1818 (1976).
Brosset, C., Andreasson, K. and Ferm, M. *Atmos. Environ.* **9,** 631 (1975).
Dzubay, T. G., Stevens, R. K. and Peterson, C. M. (1977), in *X-Ray Fluorescence Analysis of Environmental Samples* (Edited by Dzubay, T. G.), pp. 95–105. Ann Arbor Science Publishers Inc., Ann Arbor/Michigan.
Ferm, M., *Atmos. Environ.* **13,** 1385 (1979).
Gregory, G. and Moyer, R., *Rev. Sci. Instrum.* **48,** 1464 (1977).
Junge, C. and Scheich, G., *Atmos. Environ.* **3,** 423 (1969).
Klockow, D., Jablonski, B. and Niessner, R. *Atmos. Environ.* **13,** 1665 (1979).
Loo, B., Jaklevic, J. and Goulding, F., in *Fine Particles: Aerosol Generation, Measurement, Sampling and Analysis* (Edited by Kiu, P. Y. H.) pp. 311–350. Academic Press, New York.
Lippmann, M., in *Fine Particles: Aerosol Generation, Measurement, Sampling, and Analysis* (Edited by Liu, B. Y. H.), pp. 287–310. Academic Press, New York.
Mallant, R., Gouman, J. and van de Vate, J. (1978) Proceedings of the 6th Annual Conference of GAeF, Vienna, Sept. 26–28, pp. 119–126.
Marlow, W. and Tanner, R., *Anal. Chem.* **48,** 1999 (1976).
Marple, V. and Chien, C., *Environ. Sci. Technol.* **14,** 976 (1980).
McFarland, A., Ortiz, C. and Bertch, R., *Environ. Sci. Technol.* **12,** 679 (1978).
Niessner, R. (1981), Ph.D. Thesis, University of Dortmund, Dortmund, FRG.
Niessner, R. and Klockow, D., *Int. J. Environ. Anal. Chem.* **8,** 163 (1981).
Niessner, R. and Klockow, D., *J. Aerosol Sci.* **13,** 175 (1982).
Pack, M., Hill, A., Thomas, M. and Transtrum, L., *Soc. Testing Mat. Spec. Techn. Publ.* **281,** 27 (1959).
Plomp, A. (1981), Unpublished Report, 12/81, ECN Petten, The Netherlands.
Rao, A. (1975). An experimental study of inertial preseparators. Ph.D. Thesis, University of Minnesota, Particle Technology Laboratory, Minneapolis, USA, Publ. No. 269.
Slanina, J., v. Lamoen-Doornenbaal, L., Lingerak, W., Meilof, W., Klockow, D. and Niessner, R., *Int. J. Environ. Anal. Chem.* **9,** 59 (1981).
Tanner, R., Marlow, W. and Newman, L., *Environ. Sci. Technol.* **13,** 75 (1979).
Weinstein, L. H. and Mandl, R. H. (1971), in *VDI-Berichte Nr.* 164, pp. 53–63. VDI-Verlag, Düsseldorf.
Willeke, K. and Pavlik, R. (1980), in *Generation of Aerosols* (Edited by Willeke, K.), pp. 427–440. Ann Arbor Science Publishers Inc., Ann Arbor/Michigan.