Size Distribution of Aerosols From a Kraft Mill Recovery Furnace

JOHN C. BOSCH, MICHAEL J. PILAT, and BJORN F. HRUTFIORD

Abstract: The size distribution of particulates emitted from a kraft mill recovery furnace has been measured using a source test cascade impactor. This cascade impactor is designed for isokinetic sampling directly inside the stack. The particle size distributions were calculated from the weight of solids deposited on each impactor stage. The particle mass mean diameter was 1 μm at the electrostatic precipitator inlet and 1.4 μm at the outlet. Analyses for chloride and sodium as a function of particle size showed that the sodium chloride concentration is higher in the submicron particles at the electrostatic precipitator outlet but independent of size at the inlet.

Keywords: Sulfate pulping · Air pollution · Exhaust gases · Sodium chloride · Particles · Particle size distribution · Recovering · Recovery furnaces · Precipitators · Cascade impactor* · Efficiency

KRAFT mill recovery furnaces generally lose some chemicals as particulate material in the stack gases, and a variety of methods are used to control these losses, including electrostatic precipitators, wet scrubbers, etc., which operate with varying degrees of efficiency. Control of particulate emissions has always been an economic necessity but it has also become an increasingly important problem from the viewpoint of air quality. Many government agencies have recently enacted emission and air quality standards that will undoubtedly become more stringent. This legislation is directly influencing the costs of air pollution control in the pulping industry.

The size distribution of particles suspended in gases from the recovery furnace must be known before an optimum dust-removal system can be designed. Furthermore, operating costs are inversely related to the dust-collection efficiency which, in turn, depends on the sizes of particles entering the gas-cleaner. The size distribution of the outlet fume will also affect the amount of solids that "fall out" in the plant vicinity compared to the quantity remaining suspended in the air. Physical and chemical characteristics of the recovery fume as a function of the size distribution are also important criteria in designing an optimum dust-collection system. This type of information is needed in the design and evaluation of various control units and also to give the relationship of source emissions to air pollution problems caused by the mill.

Very little published information exists on the size distribution of particulate matter emitted from recovery furnaces. Day (*1*) states that most particles are less than 0.5 μm in diameter and Murdock (*2*) reported 85% of the particles to be under 1 μm in diameter although his method of measurement was not discussed. Walker (*3*) obtained samples on filters and sized the deposited particles with a microscope. His results showed 50% of the particles to be less than 1.4 μm in diameter on a number-count basis. Okita (*4*) also used a microscope to compare the sizes of recovery furnace particles with that of Na_2SO_4 in the atmosphere and counted giant particles as large as 2000 μm in diameter. Besides not providing data easily usable by design engineers, the previous work has not resulted in a routine simple method of determining size distributions of the recovery fume in individual pulp mills.

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OBJECTIVES

The main objectives of this research were to measure the size distribution of particles in exhaust gases from a kraft recovery furnace and to evaluate a new type of cascade impactor that promised a routine way to obtain size distribution information. Secondary objectives were (1) to ascertain the chemical composition of aerosols at the inlet and outlet of an electrostatic precipitator in a kraft recovery process, and (2) to determine the collection efficiency of the precipitator as a function of particle size.

RESULTS AND DISCUSSION

Cumulative weight percentages for each stage were arithmetically averaged and the combined size distributions are shown in Fig. 1. Table I summarizes the Na and Cl content of the fume as a function of particle size. All chlorides were assumed to exist as NaCl.

General

The special advantage of a cascade impactor is that particulates deposited on each stage can be individually analyzed and a tedious particle count is unnecessary to obtain the size distribution. Impactor information is also more useful than data based on a particle count because collection efficiency is usually based on particle inertia and an impactor separates particles according to inertial characteristics. By sampling with an impactor and weighing the solids deposited on each stage, a large number of small particles can be lost in sampling without greatly changing the mass distribution. An impactor also overcomes another disadvantage of counting particles where extremely short sampling periods are required to collect a usable sample on the filters.

Size distributions are defined by two parameters: (1) the intercept of the curve with the 50% probability (mass mean particle diameter) and (2) the polydispersity factor or geometric standard deviation defined as:

$$\sigma = \frac{\text{particle diameter at 50\% probability}}{\text{particle diameter at 15.87\% probability}}$$

Figure 1 shows that 50% of the outlet fume weight is composed of particles smaller than 1.4 μm in diameter. Fume from the precipitator inlet contains somewhat smaller particles since 50% of the weight is composed of particles smaller than 0.9–1.1 μm in diameter. The polydispersity of each curve indicates that the inlet fume is more monodisperse than the outlet fume. (A completely monodisperse aerosol has a polydispersity factor of 1 and would appear as a horizontal line on a log-probability chart.)

Electrostatic Precipitator Efficiency

Dust-collection efficiencies of the electrostatic precipitator could be estimated because the size distributions of particles at inlet and outlet were available and dust loadings at the inlet were known. Mass loadings at the inlet (W_I) and outlet (W_O) were calculated as:

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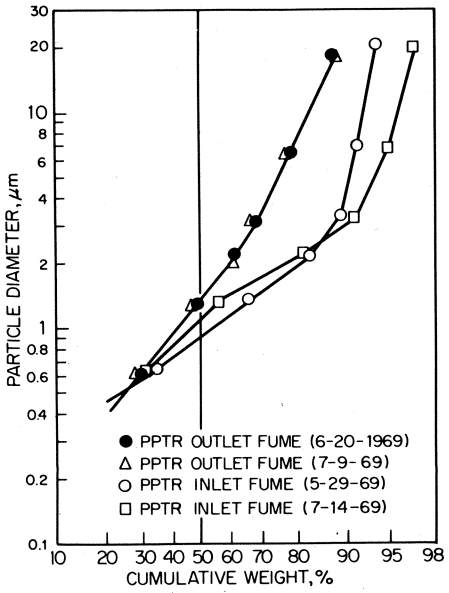

Fig. 1. Average size distribution of particles at inlet and outlet of electrostatic precipitator.

Table I. Chloride and Sodium Content of the Recovery Fume

Date	Plate	D_p, M	Dry wt., mg	Wt. NaCl, mg	Wt. Na, mg	NaCl %
June 20, 1969	1	...	18.2	2.0	3.0	11.0
(Outlet)	2	18.7	13.2	2.1	2.5	15.9
	3	...	14.7	...	...	...
	4	3.1	9.2	1.3	2.1	14.1
	5	2.2	20.0	2.9	4.4	14.5
	6	1.3	26.5	5.3	5.4	20.0
	F	0.6	43.1	9.0	8.6	20.9
July 9, 1969	1	...	18.9	2.1	5.1	11.1
(Outlet)	2	18.6	18.2	2.5	6.0	13.7
	3	6.4	15.9	2.4	5.7	15.1
	4	3.1	8.5	1.7	3.5	20.0
	5	2.0	16.8	2.7	5.9	16.1
	6	1.3	26.9	4.3	8.9	16.0
	F	0.6	33.6	7.2	11.1	21.4
July 14, 1969	1	...	16.8	1.7	6.7	10.1
(Inlet)	2	19.8	9.5	1.0	4.6	10.5
	3	6.8	21.2	2.0	9.4	9.4
	4	3.3	57.9	5.3	19.7	9.2
	5	2.2	163.8	16.5	41.1	10.1
	6	1.3	148.6	15.0	38.8	10.1
	F	0.6	207.8	24.0	46.2	11.5

$$W_I, W_O = \frac{\text{total quantity of sample}}{\text{standard dry cubic feet drawn through the meter}}$$

Incremental efficiency is defined as:

$$N^i = 1 - \left[\frac{W_O{}^i}{W_I{}^i}\right]$$

and the overall collection efficiency is:

$$N_T = \frac{\sum W_I{}^i N^i}{\sum W_I{}^i}$$

Figure 2 shows precipitator efficiency as a function of particle diameter. The plots show that collection efficiency begins to decrease for particles 1.5–2.0 μm in diameter. One reason for this is that there is a net "generation" of large particles in the precipitator which are re-entrained in the gas stream by the electrode rapping. Larger particles rather than small ones tend to be re-introduced into the flue gas (5). It must be emphasized that the apparent low collection efficiency for large particles is not because the precipitator fails to collect them from the air stream. Instead, most particles entering the precipitator are initially collected on the electrodes but large pieces of material may break away from the deposits and be recollected in the exit gases.

The annual plant cleaning program at the Tacoma mill afforded an opportunity to investigate the effect of maintenance on collection efficiency. One curve in Fig. 2 represents the "pre-cleaning" efficiency while the other is the collection efficiency after the maintenance program was completed. The plant maintenance program increases the salt cake recovery from 94.9 to 96.2% on a weight basis. This improvement is caused, in part, by the reduction in carbonaceous material entering the precipitator. (Prior to cleaning, the inlet gas stream contained a large amount of black particles. These particles were not evident after the maintenance was completed. Very little black material was observed in the outlet fume at any time.)

Composition of Collected Particulates

Analysis of material collected on each impactor stage revealed two important characteristics of the recovery fume: (1) Carbonaceous ash particles in the inlet fume are much larger than sodium-salt particles, and (2) NaCl in the outlet fume is concentrated in the small submicron particles.

Solids collected at the outlet were completely white except for the particles deposited on the top impactor plate which were somewhat "grayish." There was a quantity of black ash in all samples taken at the inlet to the precipitator. Moreover, each stage of the impactor collected varying amounts of this black ash which allowed a rough, visual estimate to be made of the fraction of black material deposited in each impactor stage.

These visible, black-white differences between samples collected on each impactor stage indicate that there are at least two independent aerosols in the recovery fume: black ash and sodium salt. This might explain why the curves in Fig. 1 are nonlinear; they reflect a sample containing multiple aerosol systems, each with a different size distribution. (Many single aerosols have log-normal size distributions which appear linear on the log-probability chart.) A qualitative separation of the inlet fume into black and white material resulted in two size distributions shown in Fig. 3 which can be compared to the single combined distribution in Fig. 1.

Sodium chloride in the outlet fume is concentrated in the smaller particles as shown in Fig. 4. NaCl content of the inlet fume, however, seems to be independent of particle size. This suggests that: (1) NaCl particles follow the same generation and growth pattern as other particulates in the fume, and (2) the high content of NaCl in submicron particles in the outlet fume may be due to a reduced tendency for NaCl to agglomerate in the precipitator. Numerical data from the NaCl analysis are too few for a definitive interpretation but it seems that the variation of NaCl with particle size is significant and that further research in this area would be useful.

If the fume is assumed to contain only NaCl and Na_2SO_4, the total expected sample weight can be determined by using the total sodium and chloride analyses. In all cases the calculated weight of each sample was less than the measured weight which implies that either (1) hydrated moisture was not completely removed from the desiccated samples, or (2) compounds other than Na_2SO_4 and NaCl are present in the fume. The difference in weights may be caused by the presence of $NaHCO_3$, or $NaCSO_3$, in the recovery fume.

CONCLUSIONS

A relatively easy method for determining size distributions of particles from kraft pulp mill recovery furnaces has been developed that involves collecting samples in a cascade impactor and weighing the solids deposited on each stage. Particle sizes in this study ranged from less than 0.6 to larger than 20 μm. This technique of size separation enables different size increments of particles to be physically and chemically analyzed.

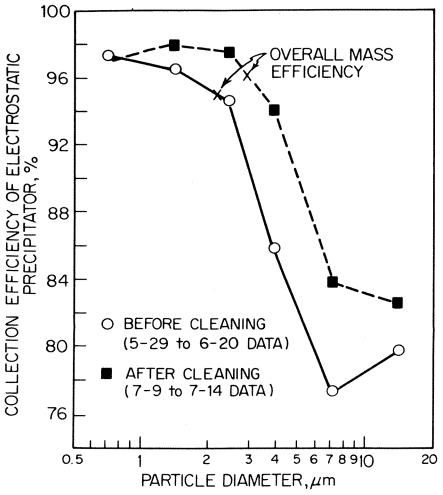

Fig. 2. Collection efficiency of electrostatic precipitator as a function of particle size.

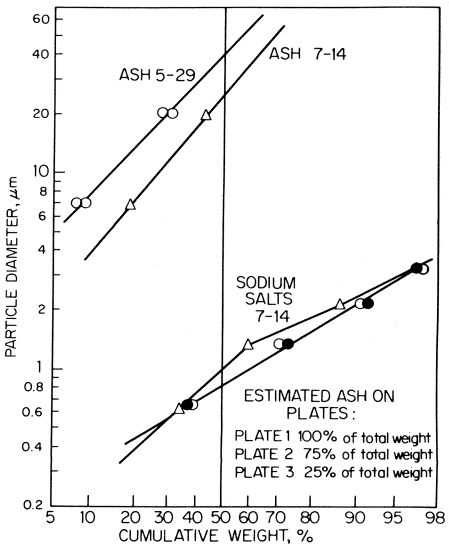

Fig. 3. Qualitative separation of inlet samples into two components—ash and sodium salts.

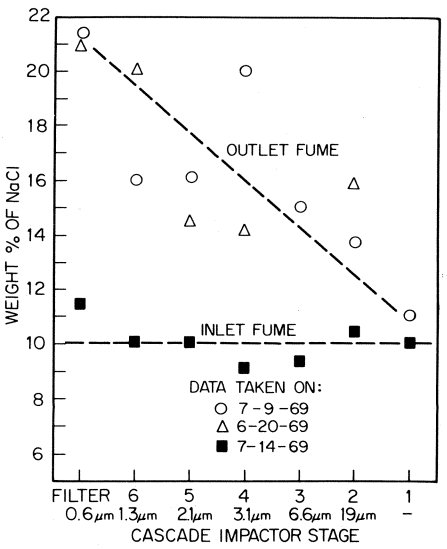

Fig. 4. NaCl content of recovery fume as a function of particle size.

Samples of the recovery fume were collected at the inlet and outlet of an electrostatic precipitator. Size distributions of particles taken at the outlet showed 50% of the weight to be composed of particles smaller than 1.4 μm in diameter. The polydispersity factor is 4.2. Particles in the inlet fume are smaller with 50% of the weight being composed of particles under 0.95–1.1 μm in diameter. Fume at the precipitator inlet also contains black, carbonaceous particles that are generally larger than the white, sodium-salt particles. Most, if not all, of this unburnt material is collected by the electrostatic precipitator and recycled to the recovery furnace.

The overall collection efficiency of the electrostatic precipitator is related to particle size and decreases rapidly for particles greater than 2.5 μm in diameter. This phenomenon is probably due to the generation of large aggregates inside the precipitator by electrode rapping.

Chloride and sodium contents of precipitator inlet and outlet fumes were determined as a function of the particle size. Sodium chloride in the outlet fume ranged from 11% by weight in the largest particles (D_p = 20 μm) to 21% in the submicron particles (D_p = 0.6 μm). Thus, NaCl is concentrated in smaller particles of fume leaving the precipitator. NaCl in the fume entering the precipitator amounts to about 10% of the total weight and appears to be independent of particle size.

EXPERIMENTAL

Test Location

A series of size distribution determinations was made at two locations within a kraft mill: (1) at a point before the electrostatic precipitator and (2) at a point prior to the gas exit from the stack. Field sampling was accomplished at the St. Regis pulp mill in Tacoma, Wash., where the following equipment was operating in the recovery process:

1. Combustion engineering recovery furnace with a designed daily capacity of 1.4 million lb of dry solids.
2. Direct-contact evaporators.
3. Research-Cottrell electrostatic precipitators.

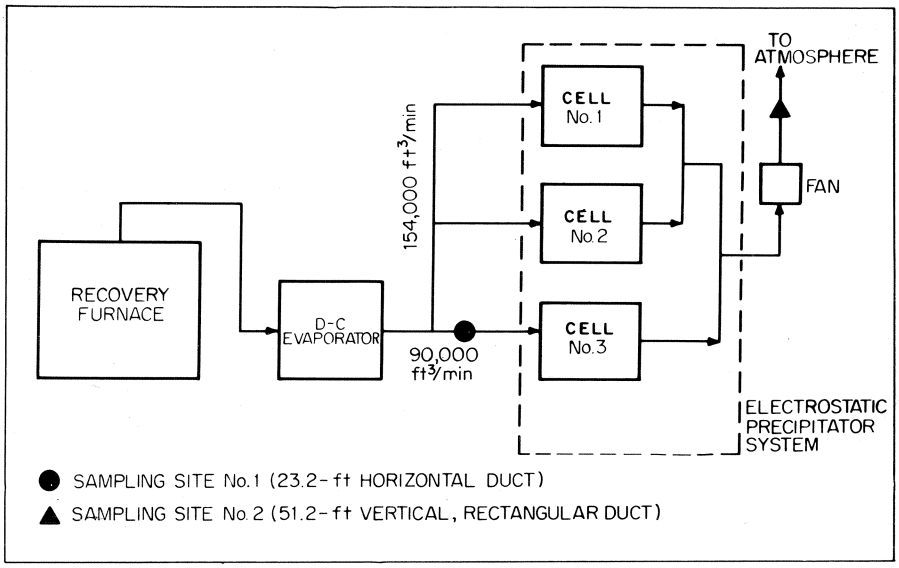

Fig. 5. Recovery system schematic showing sampling sites.

The mill has three separate recovery processes but all measurements were taken on the most modern system which is depicted in Fig. 5 along with locations of the two sampling sites.

Cascade Impactor

Particle size distributions in this study were obtained with a source test cascade impactor, the theory and design of which has been reported by Pilat, Ensor, and Bosch (5). The device is a multijet impactor that is inserted directly into the stack gases to collect the samples.

Collecting the size distribution data inside the stack using a cascade impactor has many advantages over methods involving transfer and cooling of particles or measurement by optical means. The impactor separates particulates according to their aerodynamic properties which relate directly to the design of collection equipment. Moreover, the data are obtained by simply weighing the material collected on each stage of the impactor.

The cascade impactor used is shown in Fig. 6 and consists of 3 sections: the inlet nozzle, 6 impactor stages, and the filter holder. An appropriate nozzle size is selected to attain isokinetic sampling conditions for each test. Each of the 6

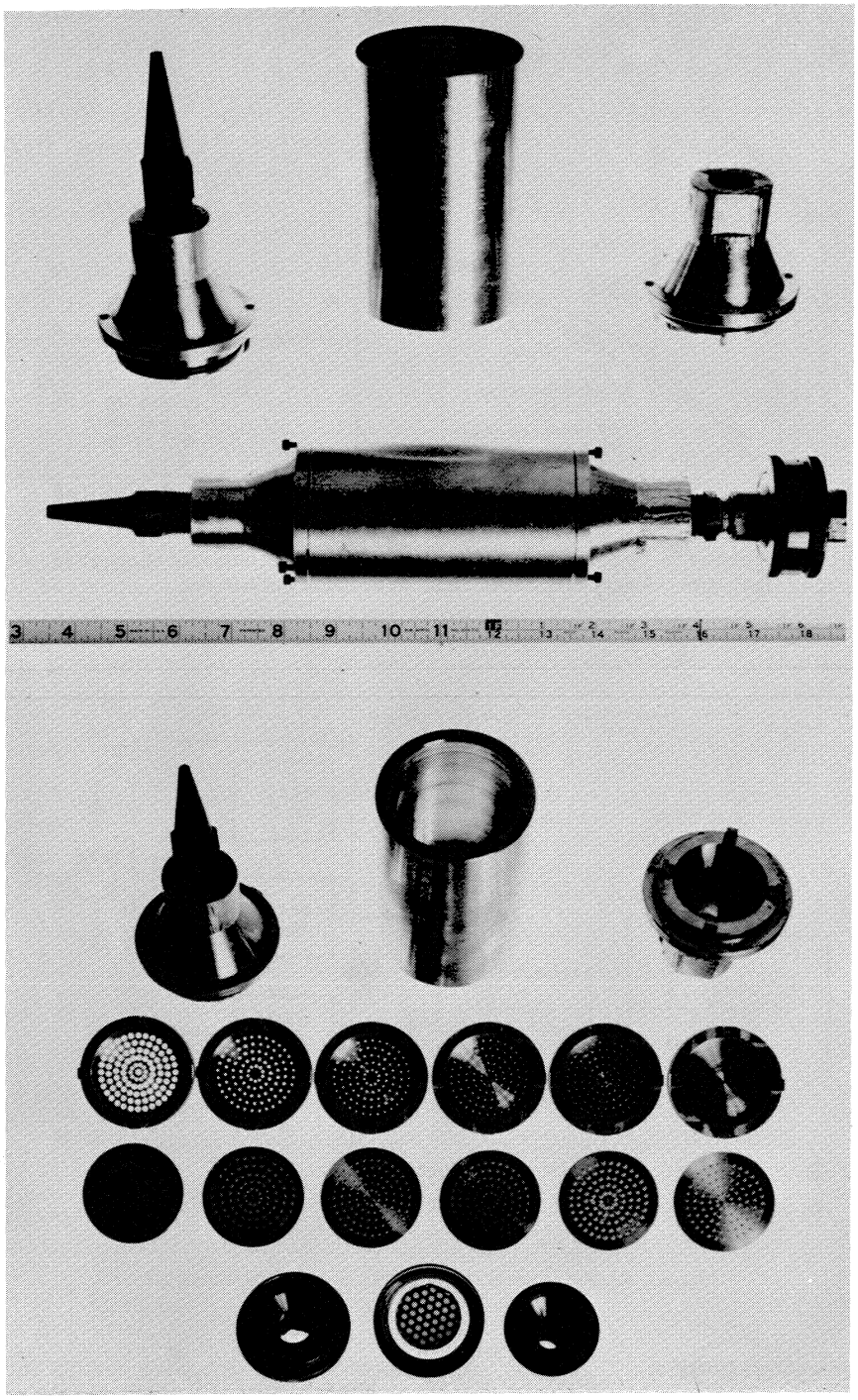

Fig. 6. Source test cascade impactor.

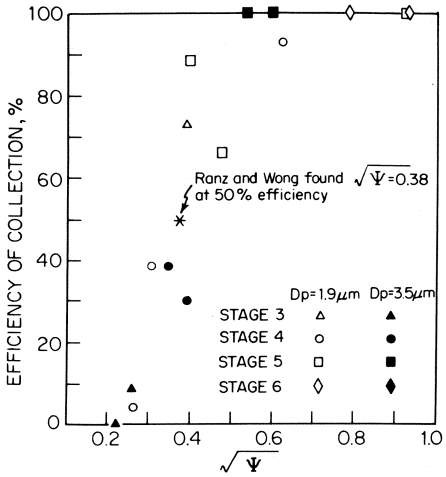

Fig. 7. Impactor collection efficiency vs. inertial impaction parameter $\sqrt{\psi}$.

stages in the main assembly is composed of an aluminum disk with 102 drilled jets together with a polished collector plate beneath the jets. The filters, located after the final impactor stage, are fabricated from the fiberglass mats usually used in high-volume atmospheric samplers.

The recovery fume contains a large range of particle sizes, and the cascade impactor used in this study was designed to separate a sample of the fume into specific particle-size increments of relatively equal weight. The ability of an impactor to do this effectively is based solely on the collection efficiency of each stage which, in turn, is a function of the inertial impaction parameter ψ. For circular jet impactors, Ranz and Wong (*6*) reported $\sqrt{\psi}$ to be 0.38 at 50% collection efficiency. The test impactor was built according to Ranz and Wong's design criteria; however, it was necessary to ascertain if the impaction parameter of this device ($\sqrt{\psi}50$) was equivalent to that found by Ranz and Wong. To do this, the impactor was calibrated using aerosols of latex-sphere particles of known diameters. The relative quantity of particles on each stage was determined under a microscope and the stage collection efficiencies were calculated for a series of particle velocities through each stage. Calibration data are shown in Fig. 7 and $\sqrt{\psi}50$ averaged 0.38 as expected.

The characteristic diameter of each stage D_p is calculated by:

$$D_p = \left(\frac{18\ \mu\ Dc}{CP_pV}\right)^{1/2} \sqrt{\psi}50$$

where:

$\sqrt{\psi}50$ = 0.38
μ = gas viscosity, p
P_p = particle density, g/cm³
V = gas velocity through jet, cm/sec
Dc = jet diameter, cm

The slip factor C is defined by Brink (*7*) as:

$$C = 1 + \frac{2L}{D_p}\,[1.23 + 0.41 \exp(-0.44\ D_p/L)]$$

where L is the mean free path of gas molecules using formula detailed by Brink. Brink considered the exponential term to be negligible when $D_p/L > 2.7$, thereby reducing the equation to $C = 1 + 2.46\ (L/D_p)$.

This simplified formula was used to calculate the slip factors which are particularly important when dealing with particles smaller than 1 μm in diameter.

In calculating the characteristic diameter for each stage, the particle density was assumed to be constant at 1.0 g/cm³ for all particle sizes. This assumption is convenient when the aerosol may contain particles of different densities. Fuchs (*8*) defined the result as a reduced characteristic diameter which is the diameter of a spherical particle of unit density and having the same falling velocity.

Auxiliary Stack Sampling Equipment

As previously discussed, the cascade impactor was inserted directly into the hot flue gases to collect the samples. After removal of all fume particles in the impactor, the clean flue gases passed

through a standard sampling train composed of two Greenburg-Smith impingers, a Rockwell dry-gas meter, and a vacuum pump. (Stack-test sampling equipment and procedures have been well documented in the literature [*9–11*].) The impingers were immersed in ice water to facilitate the condensation of moisture in the flue gases and air leakage was prevented by sealing all the metal connections with teflon tape. Velocity profiles of flue gases in the stack were obtained with an "S-type" pilot tube and inclined draft gauge.

Sampling Procedures

The following sampling techniques were developed in preliminary studies:

1. In the laboratory the top surface of each collector plate was coated with a thin layer of silicone grease. The tared and greased plates were stored in covered petri dishes until needed for assembly in the impactor.
2. Velocity profiles of flue gases were measured. A sampling point in the duct was then selected after analyzing the gas velocity distribution. All sampling points were located at least 2 ft from walls of the duct to minimize boundary effects.
3. The sampling train was assembled and 100 ml of distilled water were measured into the first impinger.
4. An appropriate nozzle size was selected and the desired meter flow rate was calculated such that isokinetic conditions could be maintained during the test.
5. The impactor was inserted into the stack and allowed to reach thermal equilibrium with the flue gases. The pump was then activated and the meter flow adjusted as needed to meet isokinetic conditions. The duration of individual tests ranged from 2 to 30 min depending on the dust loading.
6. After each test, the cascade impactor was carefully removed from the stack and taken to the laboratory for disassembly and subsequent analysis of the solids deposited on each collector plate.
7. During disassembly of the impactor, the collector plates were removed stage-by-stage and stored in covered petri dishes for reweighing.

Wall losses and dislodged particulates were evident in the impactor after sampling and ranged from zero to an estimated 30% of the total collected sample.

Some of the particles deposited on the walls of the impactor during sampling, and it was evident that a fraction of the material was also dislodged from one of the collector plates to fall into the next stage. Moreover, it was observed that some particles hit the collector plate with such force that they "bounced" onto the undersurface of the disk containing the jets. Misplaced solids due to all these causes were replaced onto the appropriate collector plate when the impactor was taken apart (i.e., wall losses before the first stage were brushed onto the first collector plate). Improvements in the design of the cascade impactor have since minimized the wall loss problems.

Sample Preparation and Analysis

All samples were conditioned to 45% RH before weighing and then were dried to 104°C and weighed again. There were usually 2 to 4 sets of valid impactor data obtained on each day of sampling at the mill. In order to collect enough material for chemical analysis, combined samples containing particles of similar sizes were made by dissolving solids collected on individual collector plates into distilled water (e.g., the deposited material on stage 5 of our test was combined with the solids on stage 5 of another test). Total sodium concentrations were determined using a Perkin-Elmer Atomic Absorption Spectrometer, Model 290B. The amount of chloride in each composite sample was determined with a Beckman specific chloride-ion electrode and a Beckman Model 1019 research pH meter.

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