

NILU  
OPPDRAGSRAPPORT NR: 46/81  
REFERANSE: 22181  
DATO: NOVEMBER 1981

A LITERATURE SURVEY OF  
METHODS FOR FRACTIONATED SUSPENDED  
PARTICULATE MATTER MEASUREMENT

BY

V. VITOLS

UTFØRT ETTER OPPDRAG FRA  
STATENS FORURENSNINGSTILSYN

NORWEGIAN INSTITUTE FOR AIR RESEARCH  
P:O.BOX 130, N-2001 LILLESTRØM  
NORWAY

ISBN-82-7247-273-2

SUMMARY

A literature review was conducted to identify methods/devices for ambient coarse and fine particulate matter measurement.

The US-type dichotomous sampler, which uses virtual impaction for particle fractionation, was judged currently the best for collecting coarse and fine particles on filters. Commercially available samplers, however, have relatively low sampling rates and may in some cases, collect only marginal particle mass for gravimetric evaluation. The EPA-type Hi-Vol sampler, equipped with the selective sampling inlet, provides samples suitable for gravimetry, but lacks a reliable and proven coarse/fine particle fractionator, and has suspected problems with artifacts.

To clarify some unsolved questions about the performance and suitability of candidate samplers, a field comparison program is proposed.

LIST OF CONTENTS

	Page
SUMMARY .....	3
1 INTRODUCTION .....	7
2 OBJECTIVE OF PROJECT .....	8
3 METHODS FOR FINE/COARSE FRACTIONATION .....	9
3.1 Methods/devices for fine fraction only .....	10
3.1.1 Impactors .....	10
3.1.2 Cyclones .....	11
3.2 Methods/devices for fine fraction - total particle samplers .....	12
3.3 Methods/devices for fine and coarse fractions ..	13
3.3.1 Impaction .....	13
3.3.2 Two-filter sampler .....	15
3.4 Inlets and sampling effectiveness .....	16
3.4.1 Vertical elutriators .....	18
3.4.2 Mixed-action inlets .....	19
4 GRAVIMETRIC EVALUATION OF FILTERS .....	24
5 DISCUSSION .....	25
5.1 Fine/coarse particle sampling methods/devices ..	25
5.1.1 Dichotomous samplers .....	25
5.1.2 SSI Hi-Vol sampler .....	27
5.1.3 Total-fine particle sampler .....	27
5.1.4 Two-filter sampler .....	28
5.2 Fine fraction only sampling methods/devices ....	28
5.2.1 Cyclone/filter sampler .....	28
5.2.2 Impactor/filter sampler .....	28
6 CONCLUSIONS .....	29
7 RECOMMENDATIONS .....	30
8 ACKNOWLEDGEMENTS .....	31
9 REFERENCES .....	31

A LITERATURE SURVEY OF  
METHODS FOR FRACTIONATED SUSPENDED  
PARTICULATE MATTER MEASUREMENT

1 INTRODUCTION

Although the various effects of airborne particles are clearly particle size-dependent, at the present time there are no ambient air quality standards or guidelines anywhere that are size-specific. For example, in Europe, the European Communities Directive 88/779/EEC (1), the OECD method (2), the West German LIB-Verfahren (3), and the British Smoke Shade method (4) require no particle size separation. All these, and the standardized Hi-Vol sampler in North-America (5,6), collect "total" suspended particles on filters, although the actual upper size cut of the sampled particles depends primarily on the intake characteristics of the sampler in question, and on the ambient wind speed.

The US EPA has moved officially towards the adoption of limited size fractionation, but the proposed revisions in the US suspended particulate matter standard are still stalled, due to political and scientific disagreements (7,8,9).

The International Standards Organization (ISO) has been deliberating size-specific particle sampling (10). Its recommendations have dealt mainly with particle size range definitions as they relate to health effects, and are advisory in nature.

In Norway the Arbeidsgruppe for luftkvalitetsstandarder of the SFT does not expect to recommend any particle fractionation until some firm action in this direction has been taken elsewhere (11).

2 OBJECTIVE OF PROJECT

The stated objective of SFT<sup>+</sup> for this project was to develop methods/equipment for routine "high volume" measurement of suspended particulate matter in the ambient air, that would allow separate collection on filters and gravimetric evaluation of fine (< 2 µm aerodynamic dia.) and coarse (> 2 µm aerodynamic dia.) fractions. The "high volume" requirement, for purposes of this study, was interpreted to mean a method/equipment that is capable of collecting sufficient sample of particulate matter, in at least 24 hours, for adequate weighing accuracy. Thus, the filter medium used must be compatible with sample size and sampling duration requirements, and preferably suitable for as many other analytical procedures (e.g., chemical, biological, microscopic), as possible.

Recent literature, relevant to airborne particulate matter fractionation methods/equipment, was reviewed. In what follows, the findings are discussed, conclusions drawn and recommendations made. For the purpose of this report, it is assumed that the reader is already familiar with the terminology and basic principles pertaining to ambient air sampling, since these will not be restated here.

---

+ Forslag fra SFT til MD angående luftforurensningsundersøkelser for 1981.

### 3 METHODS FOR FINE/COARSE FRACTIONATION

A complete characterization of ambient airborne particle distributions is probably the best for evaluation of the various effects the particles produce, but may not be cost-effective from a monitoring viewpoint. For many purposes, simple separation into two size fractions may suffice. Such procedure, normally employing particle fractionation according to their aerodynamic properties, provides information on, for example, respirable/non-respirable, accumulation mode/coarse mode, or acid/alkaline particles in the sample. A 2  $\mu\text{m}$  aerodynamic diameter cutpoint is the commonly assumed, approximate "boundary" between accumulation mode and coarse particles, and roughly separates acid and alkaline particles as well.

In addition to the restriction imposed by weighing requirements, the choice of methods/equipment for the actual separation of the coarse fraction particles from the fine depends on whether the coarse fraction must be recovered and evaluated, or not. If not, the selection of options is fairly straight-forward. If, however, the coarse fraction too is to be evaluated by filter weighing, the choice is narrowed to virtual and physical impaction, or the "two-filter" techniques. In most cases, the relative amounts of the collected coarse fraction (and thus the "total" particles as well) depend on factors such as the sampler inlet geometry, ambient wind speed, particle loading, and the actual aerodynamic size spectrum of the sampled particles (12,13). Thus, if the coarse fraction is also of interest, it is necessary to specify an upper size cut for the coarse fraction, and to employ a sampler inlet configuration with the necessary intake effectiveness<sup>+</sup> and insensitivity to wind speed changes. Although the clear intent of SFT is to evaluate both fractions, for the sake of completeness, methods/devices for fine particles only will also be briefly reviewed.

---

+ Experimentally determined intake or sampling effectiveness is defined as the percent of particles reaching the collection zone of the sampler, compared to results obtained by isokinetic sampling in the wind tunnel (where tests are conducted).

### 3.1 Methods/devices for fine fraction only

To collect only the fine fraction on filters, an aerodynamic size fractionator can be used to separate the coarse fraction from the total particle sample. Cyclones, adhesive-coated impaction stages, and horizontal elutriators fall into the category of coarse particle separators. For all, quantitative recovery of the coarse fraction is at best uncertain and often impossible. Although horizontal elutriators have been utilized as non-respirable particle pre-separators in British-design samplers for occupational exposure evaluations, the sheer size of the device to provide a 2  $\mu\text{m}$  cut at any reasonable ambient air sampling rate eliminates them from further consideration.

#### 3.1.1 Impactors

Due to their relative simplicity and sharp particle size cut-off characteristics, impactors (particularly in the "cascade" or multi-stage configuration) have been quite extensively used (although not normally for routine monitoring) to obtain mass or chemical component distributions by particle size. The performance of physical impactors (i.e., those impacting particles on solid surfaces) can be accurately predicted from recently improved theory, and it is now possible to design an impactor for a specific flow rate and be fairly certain of the particle cut-off size (14). Evidence of non-ideal operating characteristics (14,15) has, however, impaired their straight-forward application. Coarse particle bounce, which distorts the fractionated mass distributions (16), appears to be difficult to avoid even under seemingly favourable conditions (17). The use of adhesive coatings, such as oil or various greases and jellies, on impaction substrates or surfaces appears to be effective (18), provided overloading of impaction surfaces (e.g., due to high concentrations or long sampling periods) can be avoided.

Although impactors can be used in any orientation, vertical, downward-facing position for coarse particle pre-separation has been most often reported (19, 20).



### 3.1.2 Cyclones

Cyclone separators are simple to operate and only moderately complicated to construct. Small diameter, low flow rate cyclones have been used extensively in North-America as non-respirable dust pre-separators for sampling occupational environments. Ambient air sampling applications are also reported, using either specially designed (21) or commercially available cyclones (22,23) for coarse particle removal. Unfortunately, a satisfactory theory for predicting cyclone performance is still lacking, and the empirical nature of cyclone design has been a handicap. Although cyclones can be scaled to any desired flow rate, each must be calibrated to ascertain its size cut-off characteristics (24). A medium flow rate (up to 28 L min<sup>-1</sup>) cyclone, of improved design (21), is shown in Figure 1. At the higher flow rates, its cut-off curves are comparable in sharpness to those of most impactors. A few cyclones of lesser cut-off sharpness, but higher capacities, are commercially available. (For example, the 1-inch UNICO 240 Model provides a 2 μm cut at ca. 115 L min<sup>-1</sup>, and the AEROTEC 2 at ca. 1 m<sup>3</sup> min<sup>-1</sup> flowrates). Although compact in size, cyclones of conventional design are more difficult (than impactors) to incorporate in larger air samplers, because of the normally right angle inlet-outlet orientation.

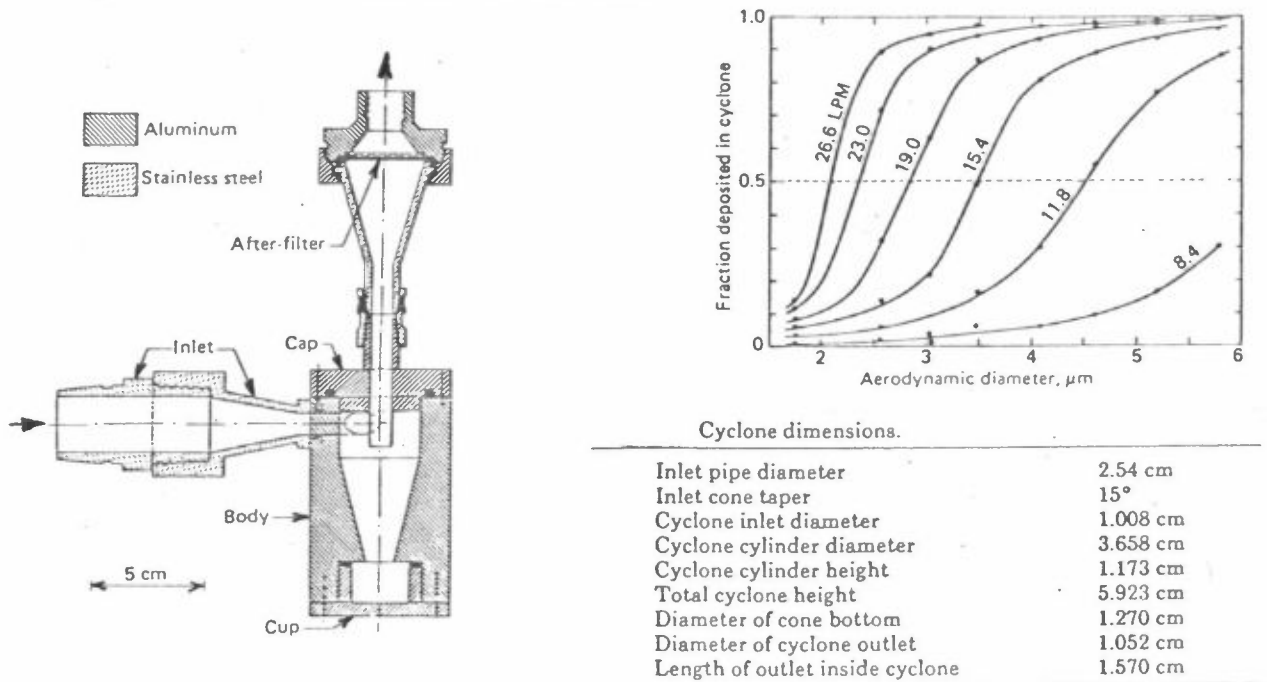


Figure 1: Cyclone assembly, dimensions and performance (21).

### 3.2 Methods/devices for fine fraction - total particle samplers

For strictly gravimetric evaluations, the mass of the coarse fraction particles can be determined by difference by using a parallel arrangement of a total and a fine fraction particle collector (21,23,24). One such parallel sampling setup is shown in Figure 2.

To ensure comparability of the measured total and calculated coarse particle measurements, and appropriate inlet for the sampler is necessary (cf. Section 3.4). An upper particle size cut-off, provided by such an inlet, can be expected to reduce the uncertainty in total (and coarse) particle mass measurements, caused by the sampling of statistically few large but heavy particles. Weighing errors can seriously impair the accuracy of the calculated coarse particle concentration obtained by this method. Further, it must be pointed out, that the total sample obtained in this manner, and consisting of a mixture of coarse and fine particles, might undergo chemical or biological alterations and thus may not represent the real nature of the coarse and fine fractions.

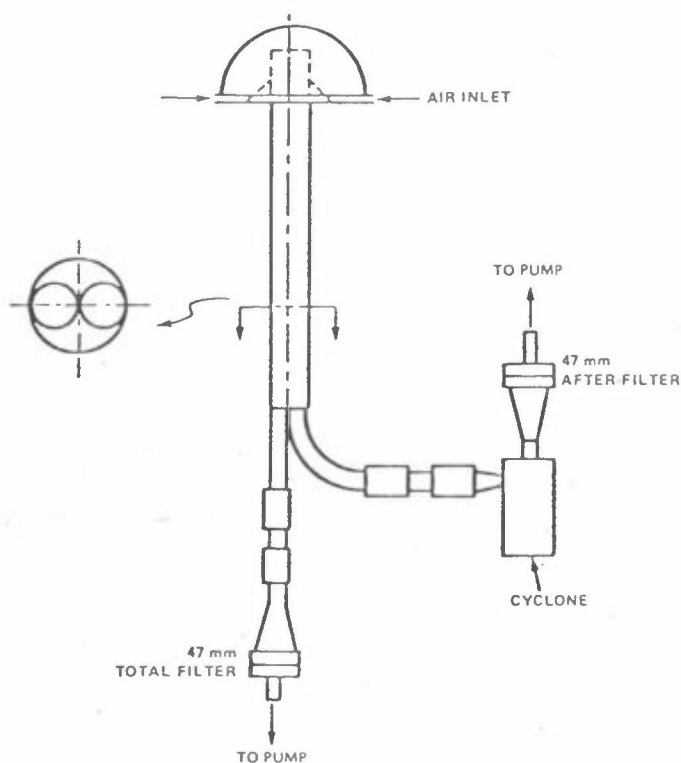


Figure 2: Assembly for airborne particle sampling with a total filter and cyclone/fine filter in parallel.

### 3.3 Methods/devices for fine and coarse fractions

If both fine and coarse fractions must be deposited on filters separately, then with presently available methods/devices the choice is limited to physical and virtual impaction, or the "two-filter" technique.

#### 3.3.1 Impaction

Although coarse fraction particles, impacted on filter substrates, would satisfy the filter collection requirement, bounce-off and reentrainment from uncoated filter substrates in physical impaction can be sufficiently severe (22) to warrant rejection of this method, unless viable remedies for this problem are found. The use of sticky coatings on impaction substrates or surfaces has been successfully used with low volume cascade impactors (e.g., 18), but has not been reported thus far for high volume impactors in routine use. The adhesive material and coating procedure must be carefully selected to suit analytical environmental (e.g., ambient temperature) requirements. Preliminary tests at NILU (unpublished data) have shown that high volume cascade impactor (Sierra) glass fibre substrates, with Apiezon grease coating<sup>+</sup>, have good tare weight stability for gravimetric evaluation. As of this writing, however, no field experience with high volume impactors using adhesive-coated substrates has been reported in the literature. At the present time, there are no impaction stages, specifically designed for coarse/fine particle fractionation, commercially available. If a stage from an existing cascade impactor is used to provide the required 2  $\mu\text{m}$  dia. cut, it is likely to be the 2nd or 3rd stage. The intake characteristics for coarse particles for these middle stages of cascade impactors are completely unknown, and there is evidence that not all of the coarse particles reach the impaction substrate. Instead of following the air stream into the impaction

---

<sup>+</sup> Apiezon L grease dissolved in toluene, and applied according to the procedure described by Cahill et al. (32).

jets, some have been observed to deposit on the impaction jet plate itself (17,25) and thus become lost from the coarse fraction.

The so-called "dichotomous" samplers of US-design employ "virtual" impaction (i.e., particle impaction into a slowly-pumped tube) to separate and collect coarse and fine particles on filters (e.g., 26). Virtual impactors do not suffer from coarse particle bounce or reentrainment, but some wall losses (particularly for liquid particles) are intrinsic to the construction features of the device. These, however, have been minimized by careful design and instrument assembly (26).

Dichotomous samplers are significantly more complicated than cyclones and physical impactors, requiring precise balance of the coarse and fine particle flows. Consequently they are more expensive, and can be more prone to operator errors. Automated versions, however, are also available with up to 20 filter pair changes in unattended operation. A schematic of a dichotomous sampler separator section and a typical performance curve are shown in Figure 3.

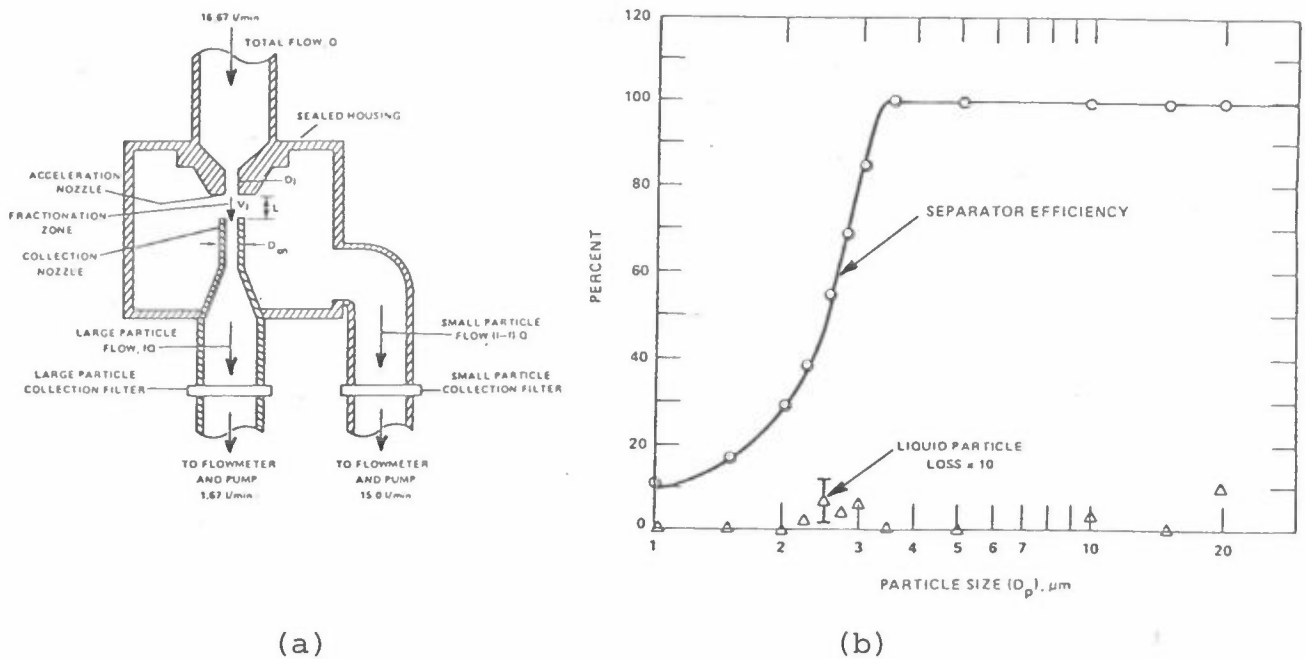


Figure 3: (a) Construction features of a dichotomous sampler; (b) Separation efficiency and wall losses.

Although some of the early dichotomous samplers of US manufacture were designed to sample at  $50 \text{ L min}^{-1}$ , only models operated at  $16.7 \text{ L min}^{-1}$  flow rate are now available.

### 3.3.2 Two-filter samplers

The "two-filter" sampler (also called the two-stage, tandem-filter, stacked filter, sequential filter sampler) utilizes a large pore size Nuclepore (NP) prefilter in series with an after-filter of appropriate characteristics (e.g., 27), which collect the coarse and fine fraction particles, respectively. The aerodynamic cut-offs of the NP filters depend on the pore size and the face velocity of the filters. The collection characteristics of nominally 8, 9.5, and 12  $\mu\text{m}$  pore diameter NP filters have been both theoretically and experimentally determined (28,29,30). The sampler can be very simple in construction, and can be assembled from commercially available components, as shown in Figure 4.

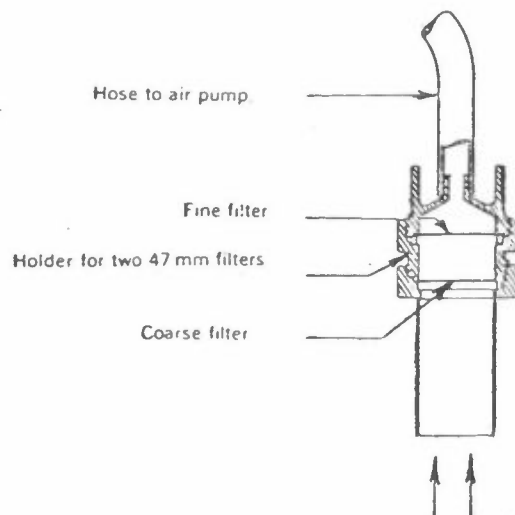


Figure 4: Sampling head of a two-filter unit.

The two-filter method allows considerable operational flexibility, since in addition to prefilter pore size, the face velocity, for a given sampling rate, can be varied by changing the effective filtration area (i.e., prefilter holder size).

As with all physical impaction, coarse dry particle bounce seems to be a problem also with the NP prefilter (22,31). Adhesive precoated NP filters are now available, which reportedly reduce the bounce-off problem, while still retaining their collection characteristics (32). There is not as yet enough information on their suitability for gravimetric evaluations. Sampling rates from 2 to 42 L min<sup>-1</sup> with two-filter samplers have been reported.

### 3.4 Inlets and sampling effectiveness

All commonly used ambient samplers have an air inlet of some sort. An ideal inlet of an ambient particulate matter sampler should allow all particles of interest to enter and be transported to the collection/sensing zone of the device, while excluding rain, snow, insects, plant matter, and other airborne debris. Research with specially designed sampler inlets, to minimize modification of particle size distributions (33), has shown that most particle mass samplers truncate the true ambient particle distributions, thereby giving concentrations less than those actually existing. If the less than perfect sampling effectiveness for a certain sampler with a given inlet geometry were constant for all conditions, then the mass collected would at least be always a consistent proportion of the true ambient concentration. Unfortunately, as recent wind tunnel tests have shown, it is also substantially affected by wind speed, particle loading, intake losses, the aerodynamic size spectrum of the particles, and in some cases even wind direction (12,34). With our present state-of-the-art sampling technology, absolute accuracy of measured airborne particle mass cannot be quantified (since truly isokinetic sampling of ambient atmosphere cannot be achieved). Size-specific and wind speed insensitive (to some specified maximum speed) inlets are then a necessity to insure a reasonable degree of equivalence between total and/or coarse particle concentrations measured by different types of particle mass samplers, or at least reproducible measurements by the same type samplers under varying wind conditions.

An omni-directional inlet of cylindrical shape (with the axis of the cylinder oriented vertically) will avoid sampling bias due to horizontal wind direction changes. Vertical elutriators, and various "mixed-action" inlets of recent US-designs all utilize the circular configuration. True horizontal elutriators (35), i.e., multi-tray or multi-tube settling "chambers", must be operated in a horizontal position and thus cannot avoid wind direction effects (without orienting wind vanes), and probably also wind speed biases.

The mixed-action inlets usually augment simple gravitational settling (the separating process in vertical and horizontal elutriators) with centrifugal and/or impaction collection to achieve (more or less) the desired intake effectiveness characteristics. There does not appear to be any sound theoretical basis for modelling the inertial effects of changing wind speed on the sampling effectiveness of such devices, and the performance of each must be assessed empirically (i.e., through wind tunnel tests).

No less important, than the above considerations for measurement comparability is the choice of particle size cut-off for the inlet. In the past, particulate matter samplers of widely different (and often unknown) sampling effectiveness have been used (e.g., 12,22,36) and efforts towards standardization were not prominent. Early proposals for revisions of the US total suspended particulate matter standard included an "inhalable particle" (IP) classification (37), concerning particles that can penetrate to the tracheobronchial and alveolar regions of the lung (9,37). A tentative nominal 50% cut-off size of 15  $\mu\text{m}$  aerodynamic diameter was first thought appropriate, and a performance "envelope" for the sampling effectiveness curves for the inlet was suggested (cf., e.g., Figures 6(b) and 6(b)). The issue of what the nominal cut-off size should be, how sharp the effectiveness curve should be, and what the permissible deviations should be became, however, subject to some controversy (7,8). As Lodge *et al.* (8) pointed out, the 15  $\mu\text{m}$  diameter cut point may be very difficult to achieve reliably in practice. It is near the maximum

of the coarse particle mode (in bimodal ambient aerosol distributions), so that even slight errors in the cut points of real inlets could lead to sizable differences in the measured coarse (and possibly total) mass measurements.

Recently the US Clean Air Scientific Advisory Committee (CASAC) recommended that the new IP standard be based on particles less than 10  $\mu\text{m}$  aerodynamic diameter instead (9). It cited two reasons for the choice: the 10  $\mu\text{m}$  cutpoint agrees better with measured values of thoracic and extra-thoracic deposition, and an inlet with 10  $\mu\text{m}$  cut-off appears to be less wind dependent and thus would contribute to more reproducible measurements.

Committee TC 146 of the ISO conducted a ballot in the spring of 1981 on whether to accept the 15  $\mu\text{m}$  or an alternate 10  $\mu\text{m}$  cut size (10). Its members decided to recommend the 10  $\mu\text{m}$  cut for standardization.

#### 3.4.1 Vertical elutriators

The basic arrangement of a vertical elutriator (sometimes called the "inverted inlet") simply consists of a vertical cylinder, aspirating upwards the air sample through its open bottom end. The theory of vertical elutriation has been revisited and further elaborated by Stöber et al. (38). Experimental evaluation of inlet effectiveness for the OECD and the LIB samplers (which use this type of inlet) has been reported by Steen and Johansson (12). The range of particle sizes used in their experiments was not wide enough to determine the 50% cut sizes, but, in general, the sampling effectiveness decreased with increased aerodynamic size of the test particles and increased wind speed. These results, taken together with Stöber et al. (38) theoretical calculations, suggest that vertical elutriators are quite sensitive to wind speed changes, unless high aspiration velocities are maintained. This, however, is not possible, because the 50% cut size itself is inlet velocity-dependent. An idea of an even simpler type of vertical elutriator, consisting of a vertical, cylindrical shroud, open at both ends, has been suggested for minimizing wind



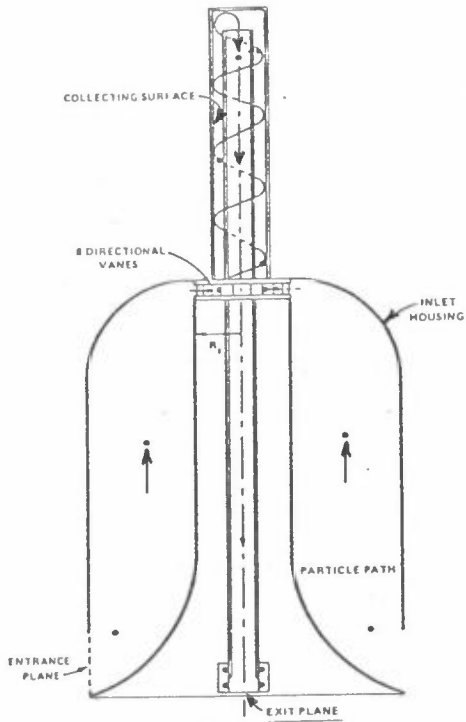
effects on inlet effectiveness (36). No studies, however, of this arrangement have been reported, and its actual performance remains unknown.

### 3.4.2 Mixed-action inlets

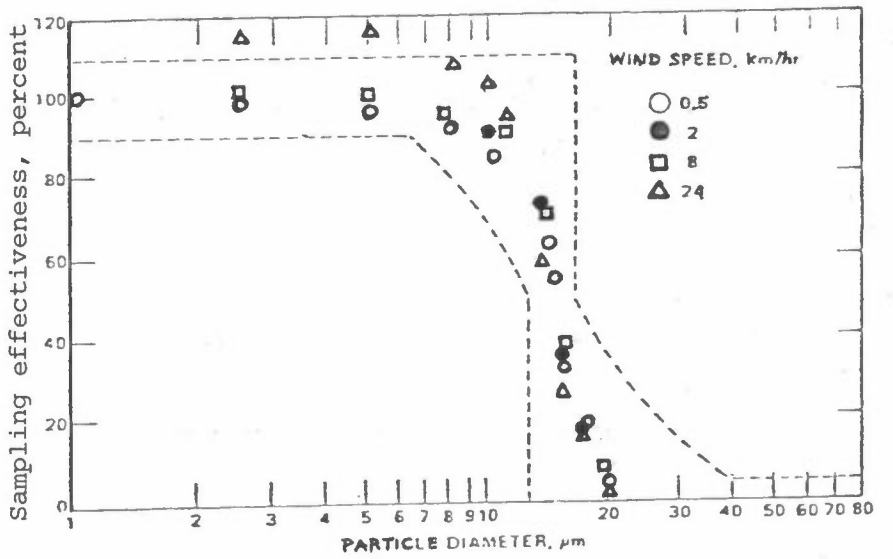
The "mixed-action" inlets, were designed and developed mainly in the USA for use with dichotomous samplers. They use vertical and horizontal elutriation in combination with cyclonic and/or impaction separation, and usually have what is called a circumferential side entrance (i.e., an annular slot). The flow patterns of air through such inlets are therefore complex, preventing reliable prediction of particle collection characteristics without wind tunnel tests.

The first inlets, developed at Texas A&M University (TAMU), were designed for sampling rates of 16.7 and 50 L min<sup>-1</sup> (39). Two inlets ("CHAMP" (40) and SSI Hi-Vol (41)), however, are capable of operating at flow rates typical (i.e., > 1 m<sup>3</sup> min<sup>-1</sup>) for US EPA-type high volume samplers.

The 15 µm cutpoint TAMU inlets later became subject to closer scrutiny (42) and were found in laboratory tests significantly affected by different wind speeds, and failing to satisfy the demands of the suggested performance envelope. Subsequently, improved 15 µm cut inlets have been laboratory (36,43) and field (44) tested. These are the Colorado State University (CSU), the University of Minnesota/Lawrence Berkeley Laboratory (UMLBL), and the Aeroenvironment (AERO) inlets. The schematic construction features and performance characteristics of the CSU and UMLBL inlets are illustrated in Figures 5 and 6, respectively. Both inlets have circumferential side entrances. (Note that the suggested performance envelopes, shown in Figures 5(b) and 6(b), are for the 15 µm IP cutpoint.)

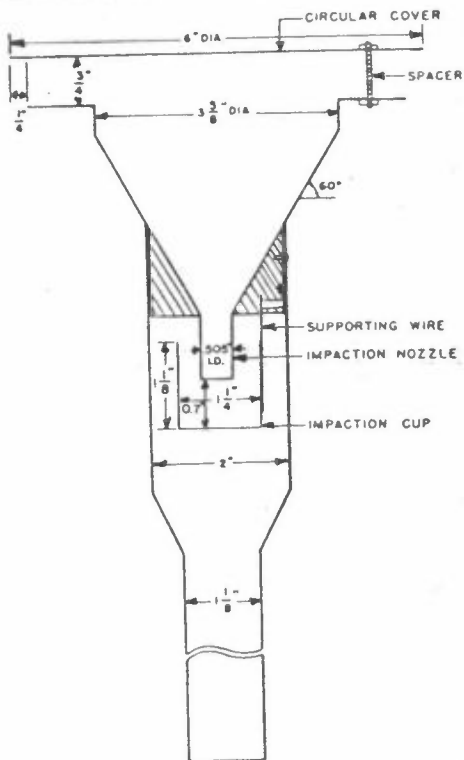


(a)

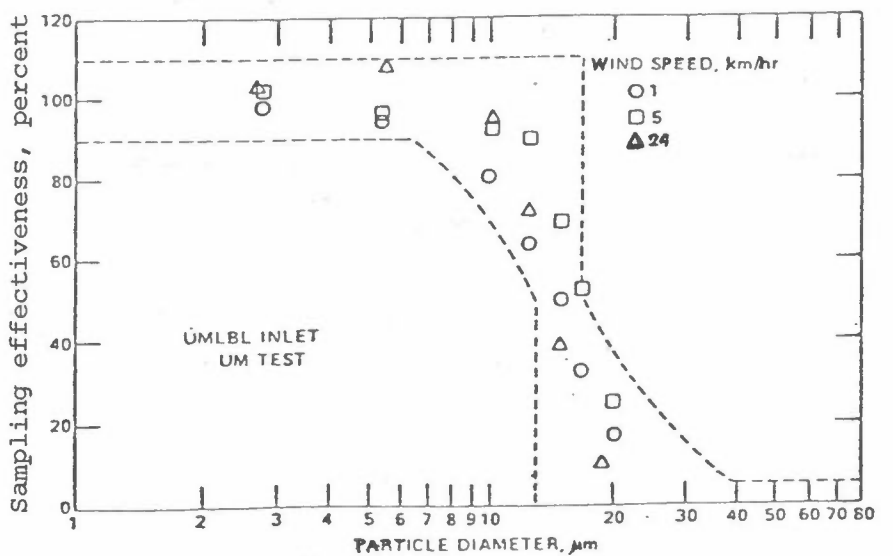


(b)

Figure 5: (a) CSU inhalable particle inlet in sectional view; (b) experimental sampling effectiveness for various wind speeds.



(a)



(b)

Figure 6: (a) UMLBL inhalable particle inlet in section view; (b) experimental sampling effectiveness for various wind speed. (The dashed lines indicate the suggested performance envelope for 15 μm diameter cut-off inlets.)

The main particle fractionation process for the CSU IP inlet is cyclonic action, which is preceded by vertical elutriation. The UMLBL inlet subjects the aspirated particles first to some horizontal elutriation and then impacts the coarser particles in a cup, which minimizes bouncing particle reintrainment and carry-over. The particle separation curves are quite sharp for both near the 15  $\mu\text{m}$  aerodynamic diameter cut-off size. Both designs are relatively unaffected by changes in windspeed (up to the maximum test speed of ca.  $7 \text{ m s}^{-1}$ ), and both have sampling effectiveness in excess of 100% for particles  $< 10 \mu\text{m}$  dia. at high wind speeds.

Specific information on the AERO inlet is not available in the open literature, but the inlet reportedly (44) consists of two concentric cylinders with offset slots. The space between the cylinders acts as a "stilling volume", and the air stream passes into a small central cylinder with a gap, that defines the particle cutpoint.

A recent study (44) attempted to determine whether differences in measurements obtained with the earlier TAMU and the improved CSU, UMLBL and AERO inlets were significant enough to invalidate past data collected with the TAMU inlet. The comparison of results in Figure 7 from TAMU and CSU inlets indicate that, under the conditions of the field trials, there was reasonably good agreement. Similar results were found also from comparisons with UMLBL and AERO. Only periods with high gusty winds resulted in poor reproducibility of coarse particle mass for TAMU inlets, confirming that under such conditions the improved inlets perform more reliably. These results, however, cannot be considered entirely typical, because of the quite low particle concentrations and a narrow wind speed range prevailing during most test periods.

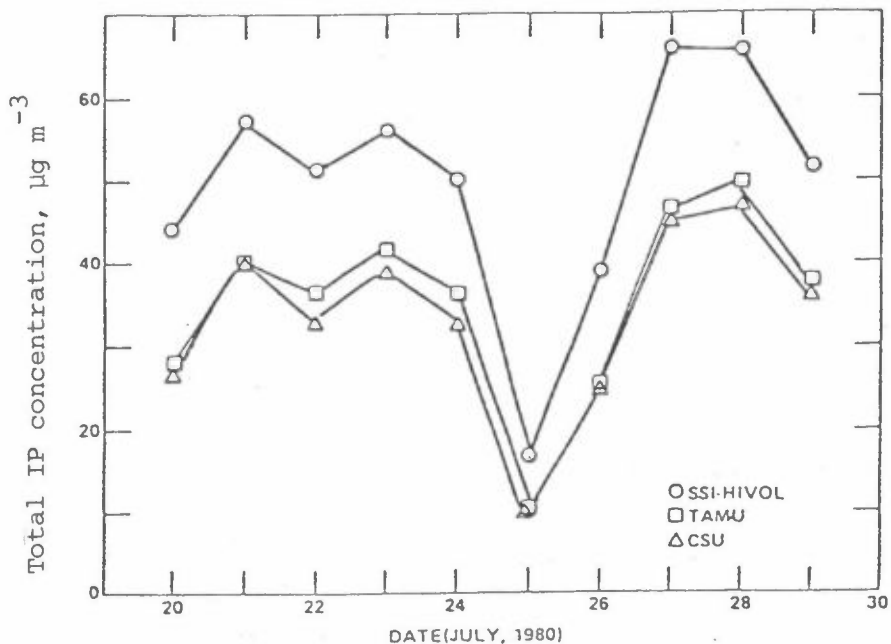
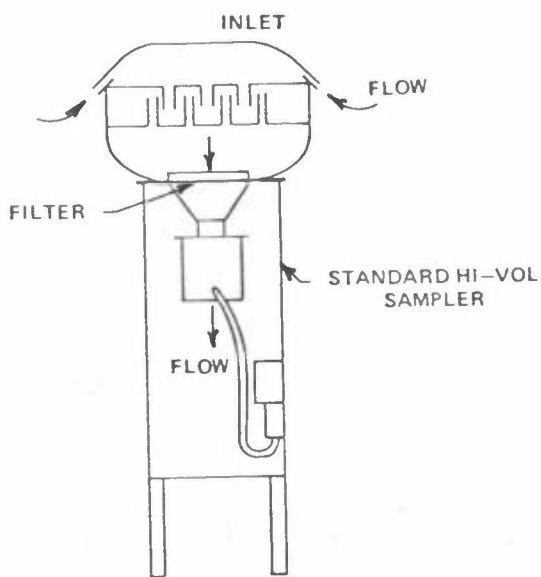
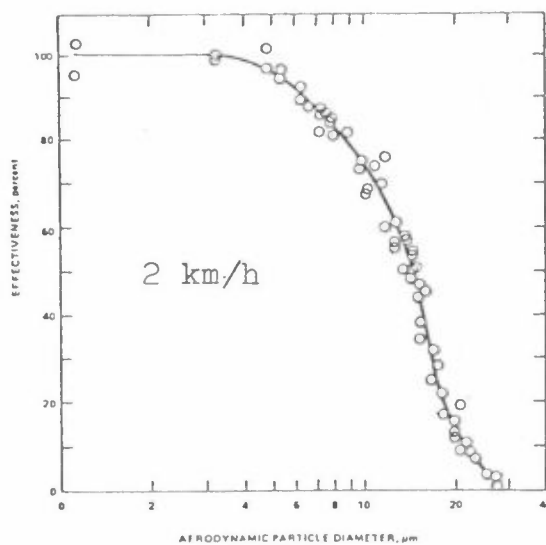


Figure 7: Total mass concentrations measured simultaneously with dichotomous samplers, using TAMU and CSU inlets, and an SSI Hi-Vol sampler (44).  
(Heavy rain during the 25 June 1980 period.)

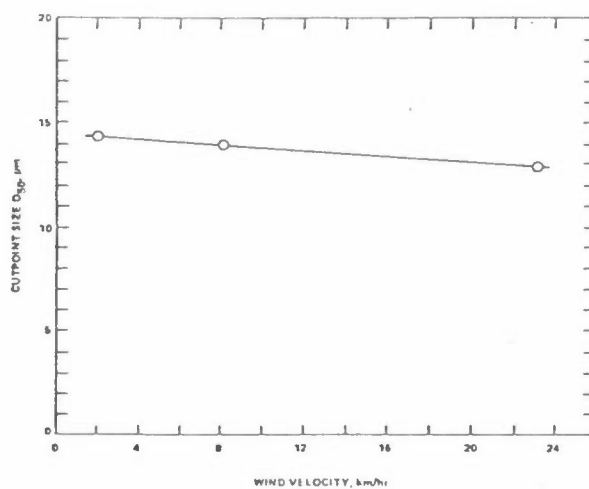
The only commercially available, high volume ( $1.13 \text{ m}^3 \text{ min}^{-1}$ ) inlet with a 50% cut size near  $15 \mu\text{m}$  is the size selective inlet (SSI) for the US EPA-type Hi-Vol sampler, shown in Figure 8. It utilizes horizontal elutriation and impaction, and is also sufficiently insensitive to wind speed changes. As can be seen in the figure, to achieve the  $15 \mu\text{m}$  cut-off at such high flow rates, the necessary dimensions of this type of inlet become rather large.



(a)



(b)



(c)

Figure 8: (a) Schematic of the SSI Hi-Vol sampler; (b) experimental sampling effectiveness of the SSI; (c) cutpoint variation with windspeed of the SSI.

A comparison of total inhalable particle mass concentrations, simultaneously collected with dichotomous and SSI Hi-Vol samplers, is also shown in Figure 7. The SSI Hi-Vol collected more mass than the dichotomous samplers. The additional mass is thought to be partly due to artifact sulphate and nitrate formation during sampling by reactions of ambient  $\text{SO}_2$  and gaseous  $\text{HNO}_3$  with the glass fibre filter of the Hi-Vol sampler (44). The Teflon membrane filters, used with dichotomous samplers, are inert to  $\text{SO}_2$  reactions, but can create a negative nitrate artifact (loss).

It can be further speculated, that internal wall losses in the dichotomous samplers (26) and/or the possibility of some re-sampling of its own exhaust by the SSI Hi-Vol (45) may have further contributed to the discrepancy.

It can be expected that all the 15  $\mu\text{m}$  inlet units will be redesigned if CASAC's 10  $\mu\text{m}$  diameter cutpoint is the choice for the new U.S. inhalable particle standard.

#### 4 GRAVIMETRIC EVALUATION OF FILTERS

Perhaps the main limitation in the selection of a candidate method/sampler for size-fractionated sampling of suspended particulate matter is the need to have enough particle mass collected during a 24-h sampling period for gravimetric evaluation. In relatively clean ambient air, the medium flow rate samplers (e.g., dichotomous and sharp cut-off cyclone samplers) will collect only sub-milligram quantities. For these, micro-balances with at least 10  $\mu\text{g}$  resolution and very careful filter handling and weighing are required for reliable weighing results. Additionally, erroneous gravimetric measurements may result from water sorption on filters and electrostatic effects of membrane-type filters. Since the latter have superior artifact formation "resistance", their choice as the filtration medium is likely, but even they have been shown to exhibit weight gains under varying relative humidity conditions (46). Furthermore, some types of membrane filters appear to carry residual charges, additional to those acquired during air sampling, and these are apparently hard to neutralize with conventional charge neutralization procedures, and require lengthy conditioning (47). All such complications are, of course, of considerably lesser importance for larger mass collections, obtained with high volume samplers, such as the EPA-type Hi-Vol.

As an alternative to microbalance weighing, beta-gauging methods can be used (48,49). They have been shown to be equivalent in accuracy to gravimetric method, require much less sample handling and conditioning, and are advantageous for larger-scale monitoring programs.

## 5 DISCUSSION

The methods/devices are discussed in order of decreasing preference (as judged by the reviewer).

### 5.1 Fine/coarse particle sampling methods/devices

The following particulate matter samplers can be used to collect both coarse and fine fractions of airborne suspended particulate matter on filter media.

#### 5.1.1 Dichotomous samplers

For the collection and evaluation of both coarse and fine particles on filters, the US-type dichotomous sampler has several advantages. Field experience with commercial manual and automatic models is accumulating (e.g., 22,44), although all the units used have had a nominal 15  $\mu\text{m}$  upper cut inlets. Prototype wind-insensitive inlets, with 10  $\mu\text{m}$  diameter cutpoint have been tested so far only in the laboratory. The anticipated adoption of the inhalable particle standard in the USA should, however, stimulate their further development and evaluation, and quicken commercial availability.

Although the dichotomous sampler is likely to be the most expensive choice, it is purchased as a complete system. In a way, it is unfortunately that the relatively low flow rate model (15  $\text{L min}^{-1}$  for the fine fraction) is the one likely to become the accepted "standard" (in preference to the earlier 50  $\text{L min}^{-1}$  version), which in areas with very low particle concentrations may provide

only marginally sufficient mass for gravimetric evaluation (cf. Section 4). Measurement reproducibility (with the same inlet) is dependent on the amount of mass collected on the filters, and on the care taken during filter handling and weighing. Figure 9 illustrates sample reproducibility obtained with TAMU inlet-equipped dichotomous samplers and beta-gauging (44). The coefficient of variation (i.e., the ratio of standard deviation to the mean) is lower for fine fraction particles and improves with larger mass loadings. The automated models are, in general, capable of better reproducibility than the manual ones (22).

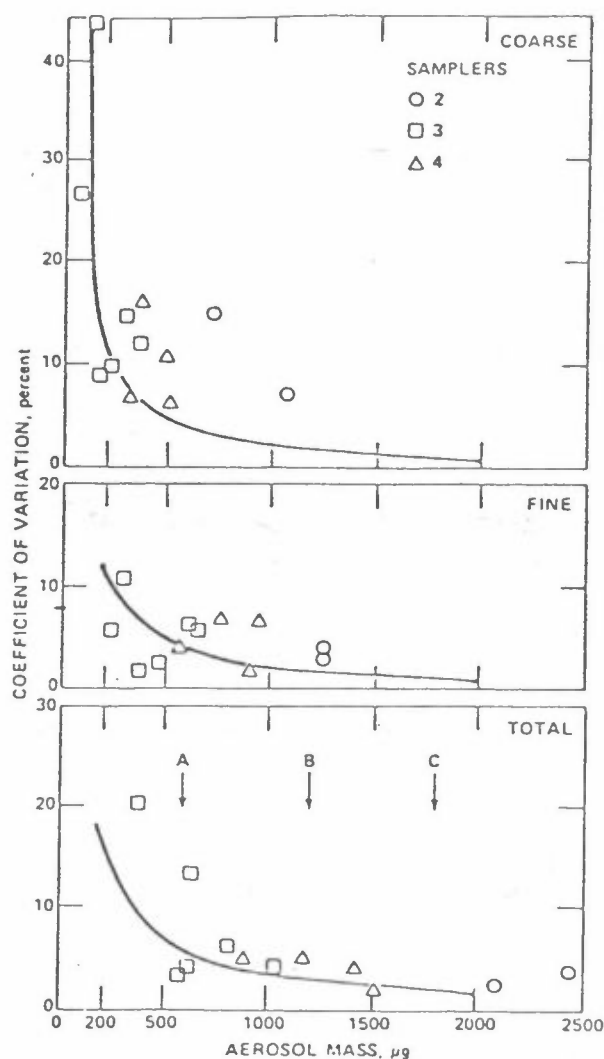


Figure 9: Reproducibility of aerosol mass (measured by beta-gauge) collected using TAMU inlets. Total collected mass corresponding to atmospheric concentrations of 20, 30 and 75  $\mu\text{g}/\text{m}^3$  and sampling periods of 24 hours are shown by A, B and C, respectively (44).



### 5.1.2 SSI\_Hi-Vol\_sampler

The US EPA-type high volume sampler is a time-proven ambient air sampler. It retains its reliable performance with the addition of the size selective inlet (SSI), but lacks a 2  $\mu\text{m}$  cutpoint fractionator.

The use of existing high volume cascade impactor stages (e.g., Sierra or Andersen) for this purpose, without adhesive coatings, appears inadvisable. Coarse particle bounce, reentrainment and carry-over (16,17,22) are likely to distort both coarse and fine particle measurements. Adhesion performance of various coating materials for impaction stage substrates under different mass loading and air temperature conditions has not been investigated.

The intake characteristics of the middle stages of commercial high volume cascade impactors, when used individually as coarse/fine particle fractionators, are also not known, and coarse particle loss from the coarse fraction sample cannot be ruled out. Artifact formation may also be a potential problem (cf. Section 3.4.2) when glass fibre filters and substrates are used.

Field comparison tests under all weather conditions are needed to assess the performance of an SSI Hi-Vol sampler, equipped with an adhesive-coated substrate impaction stage for ca. 2  $\mu\text{m}$  aerodynamic diameter fractionation (cf. Section 6).

### 5.1.3 Total-fine\_particle\_sampler

The total-fine particle sampling method appears best suited for medium flowrate applications. No commercial units are available at this time. The parallel sampling arrangement requires an upper cut inlet for the total sampler. For this dichotomous sampler inlets could be used, but this would impose a relatively low flow rate limitation and the associated weighing accuracy problems.

The use of Hi-Vol samplers in a parallel arrangement (e.g., one with SSI and another with a cyclone or sticky impaction stage) would make such a "sampler" just simply too bulky for practical routine use.

#### 5.1.4 Two-filter sampler

The two-filter sampler is promising in principle, as well as inexpensive. It allows considerable flexibility in choosing sampling rates. There is, however, not enough field experience on whether adhesive-coated prefilters can overcome coarse particle bounce. Dichotomous sampler inlet could again be used, provided flow rate limitations and weighing accuracy is acceptable. More development and testing work is needed (cf. Section 6).

### 5.2 Fine fraction only sampling methods/devices

The following samplers can be used to collect fine fraction particles only. For these, special inlets are normally not required.

#### 5.2.1 Cyclone/filter sampler

A filter sampler with a cyclone preseparator, for removing the coarse particle fraction, is relatively simple in construction and operation. It allows considerable flexibility in sampling rate selection. Cyclone separators do not overload, and particle carry-over does not normally occur. Cyclone design has been empirically improved to the point where their performance rivals that of impactors in cut-off sharpness. Commercial availability, however, is limited at the present time, but fabrication details are found in the open literature.

#### 5.2.2 Impactor/filter sampler

A filter sampler with an impaction stage preseparator is even easier to construct than a cyclone/filter sampler. Impaction

theory is now well understood, and it is possible to design an impactor for a specific flowrate to provide the desired particle cut-off with reasonable certainty. Coarse particle bounce, however, can result in large oversampling errors for the fine particle fraction. Coating of the impaction surface with sticky materials minimizes carry-over, but, in addition, overloading of the impaction stage must be avoided.

## 6 CONCLUSIONS

According to the literature review conducted for this project, it is concluded that to achieve the objective of coarse and fine fraction collection (at ca. 2  $\mu\text{m}$  aerodynamic diameter cut point) on filters and gravimetric evaluation, the following options are available. They are largely based on commercially available, or easily manufactured and assembled devices/components, and are listed in order of decreasing preference (as judged by the reviewer).

- (a) US-type dichotomous sampler (coarse and fine fractions collected on filters), with inlet of known upper size cut and performance;
- (b) high volume sampler (US EPA-type), equipped with size selective inlet (SSI) and impaction stage(s); (Coarse and fine fractions collected on adhesive-coated filter substrate and filter, respectively.)
- (c) total and fine particle (e.g., cyclone or adhesive coated impactor equipped filtering device) samplers in parallel, with inlet of known upper size cut-off and performance; (Total and fine particles collected on filters.)
- (d) two-filter sampler (coarse and fine fractions collected on filters), with inlet of known upper size cut-off and performance.

If fine fraction evaluation alone is sufficient:

- (e) cyclone preseparator and (after) filter for fine fraction collection;
- (f) adhesive-coated impactor preseparator and (after) filter for fine fraction collection.

In options (e) and (f), the samplers do not require special inlets with known cut-offs and performance.

## 7 RECOMMENDATION

Because of certain unresolved questions about options (a) and (b) (Section 6), it is recommended that a field comparison sampling program be initiated at selected sites and time periods, typical of the Norwegian environment.

The comparisons should involve parallel sampling with duplicate:

- (a) US-type dichotomous samplers;
- (b) SSI Hi-Vol samplers, modified to incorporate for the ca. 2  $\mu\text{m}$  dia. cut a commercial (e.g., Sierra Hi-Vol cascade impactor) impaction stage with adhesive-coated substrate;
- (c) SSI Hi-Vol samplers.

The aims of the comparison program should include the evaluation of:

- (i) reproducibility of inhalable fine, coarse and/or total mass concentrations, for each sampler;
- (ii) equivalence of inhalable fine, coarse and total mass concentrations measured by dichotomous and modified SSI Hi-Vol samplers;
- (iii) equivalence of total inhalable particle mass concentrations measured by dichotomous, modified SSI Hi-Vol, and SSI Hi-Vol samplers;

- (iv) best impaction stage substrate medium/adhesive material combination(s) for the modified SSI Hi-Vol sampler;
- (v) best (after) filter material(s) for the SSI Hi-Vol sampler to avoid or minimize artifact effects.

It would be beneficial to add a two-filter sampler to such an evaluation program as well.

## 8 ACKNOWLEDGEMENT

Extensive discussions with L.O. Hagen, J.E. Hanssen, St. Larssen, and H. Dovland are most gratefully acknowledged.

## 9 REFERENCES

- (1) Legislation, 80/779/EEC: Council Directive of 15 July 1980 on air quality limit values and guide values for sulphur dioxide and suspended particulates. *Official Journal of the European Communities*, 23, No L229/39, 30 August 1980.
- (2) Report of the Working Party: "Methods of measuring air pollutants", No. 17913. OECD, Paris, 1965.
- (3) Herpertz, E.: "Ein einfaches Langzeitmessverfahren zur Bestimmung der Staubkonzentration in der bodennahen Atmosphäre (LIB-Verfahren). *Staub-Reinhalt. Luft* 29, 408-413, 1969.
- (4) British Standards Institution: "Methods for the Measurement of Air Pollution. Part 2: Determination of Concentrations of Suspended Matter". B.S. 1747, Part 2, British Standards Institution, London, 1964.

- (5) National Primary and Secondary Ambient Air Quality Standards. U.S. Environmental Protection Agency. *Federal Register* 36, 8186, April 30, 1971.
- (6) "Standard Reference Method for the Measurement of Suspended Particulates in the Atmosphere (High Volume Method)." Report EPS 1-AP-73-2, Air Pollution Control Directorate, Environment Canada, Ottawa, 1973.
- (7) Kashdan, E.R. and Ranade, M.B.: "Draft. Summary of the Workshop on the Federal Reference Method for Inhalable Particulates." US Environmental Protection Agency, Nov. 1979.
- (8) Lodge, Jr., J.P., Waggoner, A.P., Klodt, D.T. and Crain, C.N.: "Non-health effects of airborne particulate matter." *Atmos. Environ.* 15, 431-482, 1981.
- (9) Hileman, B.: "ES&T Outlook. Particulate matter: the inhalable variety." *Envir. Sci. Technol.* 15, 983-986, 1981.
- (10) "Size definitions for particle sampling: Recommendations of ad hoc working group appointed by Committee TC 146 of the International Standards Organization." *Amer. Ind. Hyg. Assoc. J.* 42, A-64 to A-68, 1981.
- (11) Schjoldager, J., NILU, Personal communication. June 1981.
- (12) Steen, B. and Johansson, B.: "The sampling efficiency of two types of inlets commonly used for the sampling of aerosols in ambient air." IVL Rep. B 230, Swedish Water and Air Pollution Research Laboratory, Gothenburg, 1975.
- (13) Kolak, N.P. and Visalli, J.R.: "Comparison of three methods for measuring suspended-particulate concentrations." *Envir. Sci. Technol.* 15, 219-224, 1981.
- (14) Marple, V.A. and Willeke, K.: "Inertial impactors, theory, design and use." In *Fine Particles. Aerosol Generation, Measurement, Sampling and Analysis*. Liu, B.Y.H., Ed., Academic Press, 1976; pp 411-446.
- (15) Rao, A.K. and Whitby, K.T.: "Nonideal collection characteristics of single stage and cascade impactors." *Amer. Ind. Hyg. Assoc. J.* 38, 174-179, 1977.

- (16) Walsh, P.R., Rahn, K.A. and Duce, R.A.: "Erroneous elemental mass-size functions from a high-volume cascade impactor." *Atmos. Environ.* 12, 1793-1795, 1978.
- (17) Vitols, V.: Rural aerosol measurements with a high-volume Sierra impactor. NILU TN 16/77, Norwegian Institute for Air Research, Lillestrøm, 1977; pp 66-69.
- (18) Lawson, D.R.: "Impaction surface coatings intercomparisons and measurements with cascade impactors." *Atmos. Environ.* 14, 195-199, 1980.
- (19) Steen, B. and Johansson, B.: "An impactor to be used in connection with the LIB procedure for the determination of particle concentrations in ambient air." IVL Rep. 231, Swedish Water and Air Pollution Research Laboratory, Gothenburg, 1975.
- (20) Marple, V.A., University of Minnesota, Personal Correspondence, March 1976.
- (21) John, W. and Reischl, G.: "A cyclone for size-selective sampling of ambient air." *J. Air Poll. Contr. Assoc.* 30, 872-876, 1980.
- (22) Camp, D.C., Van Lehn, A.L., Loo, B.W., Dzubey, J.G. and Stevens, R.K.: Intercomparison of Samplers Used in the Determination of Aerosol Composition. EPA-600/7-78-118, U.S. EPA, Research Triangle Park, NC, 1978.
- (23) Bernstein, D.M., Kleinman, M.T., Kneip, Y.J., Chan, T.L. and Lippman, M.: "A high-volume sampler for the determination of particle size distributions in ambient air." *J. Air Poll. Contr. Assoc.* 26, 1069-1072, 1976.
- (24) Lippmann, M.: "Cyclone sampler performance." *Staub-Reinhalt. Luft* 39, 7-11, 1979.
- (25) Vitols, V.: Airborne sea salt mass concentration and size distribution measurements on Karmøy. NILU TN 5/76, Norwegian Institute for Air Research, Lillestrøm, 1977.
- (26) Loo, B.W., Jaklevic, J.M. and Goulding, F.S.: "Dichotomous virtual impactors for large scale monitoring of airborne

- particulate matter." In *Fine Particles. Aerosol Generation. Measurement, Sampling and Analysis*. Liu, B.Y.H., Ed., Academic Press, 1976.
- (27) Heidam, N.Z.: "Review: aerosol fractionation by sequential filtration with Nuclepore filters." *Atmos. Environ.* 15, 891-904, 1981.
- (28) Cahill, T.A., Ashbough, L.L., Barone, J.B., Eldred, R.A., Feeney, P.J., Flocchini, R.G., Goodart, C., Shadoan, D.J. and Wolfe, G.W.: "Analysis of respirable fractions in atmospheric particulates via sequential filtration." *J. Air Poll. Contr. Assoc.* 27, 675-678, 1977.
- (29) Buzzard, G.H. and Bell, J.P.: "Experimental filtration efficiencies for large pore Nuclepore filters." *J. Aerosol Sci.* 11, 435-438, 1980.
- (30) Parker, R.D. and Buzzard, G.H.: "A filtration model for large pore Nuclepore filters." *J. Aerosol Sci.* 9, 7-16, 1978.
- (31) John, W., Reischl, G., Goren, S. and Plotkin, D.: "Anomalous filtration of solid particles by Nuclepore filters." *Atmos. Environ.* 12, 1555-1557, 1978.
- (32) Cahill, T.A., Eldred, R.A., Barone, J.B. and Ashbaugh, L.L.: Ambient aerosol sampling with stacked filter units. Report No. FHWA-RD-78-178, Federal Highway Administration, NTIS, Springfield, VA., 1979.
- (33) Lundgren, D.A. and Paulus, H.J.: "The mass distribution of large atmospheric particles." *J. Air Poll. Contr. Assoc.* 25, 1227-1231, 1975.
- (34) Wedding, J.B., McFarland, A.R. and Cermak, J.E.: "Large particle collection characteristics of ambient aerosol samplers." *Envir. Sci. Technol.* 11, 387-390, 1977.
- (35) Smith, W.B., Cushing, K.M., Thomas, M.C. and Wilson, Jr., R.R.: "Some aerodynamic methods for sampling inhalable particles." In *Proceedings: Advances in Particle Sampling and Measurement*, Smith, W.B., Ed., EPA-600/9/80-004, U.S. EPA, Research Triangle Park, NC, 1980.



- (36) Liu, B.Y.H. and Piu, D.Y.: "Aerosol sampling inlets and inhalable particles." *Atmos. Environ.* 15, 589-600, 1981.
- (37) Miller, F.J., Gardner, D.E., Graham, J.A., Lee, Jr., R.E., Wilson, W.E. and Bachmann, J.D.: "Size considerations for establishing a standard for inhalable particles." *J. Air Poll. Contr. Assoc.* 29, 610-615- 1979.
- (38) Stöber, W., Holländer, W. and Morawietz, G.: Health-related sampling of total particulate matter in air with elutriating filter devices. Report (draft) of the Fraunhofer-Institut für Toxikologie und Aerosolforschung, Münster-Roxel, W. Germany, 1980.
- (39) McFarland, A.R., Wedding, J.B. and Cermak, J.E.: "Wind tunnel evaluation of a modified Andersen impactor and an all weather sampler inlet." *Atmos. Environ.* 11, 535-539, 1977.
- (40) Willeke, K. and McFeters, J.J.: "Calibration of the CHAMP fractionator." Particle Technology Laboratory Publication No. 252, Department of Mechanical Engineering, University of Minnesota, Minneapolis, MN., March 1975.
- (41) McFarland, A.R., Ortiz, C.A. and Rodes, C.E.: "The size selective inlet for the Hi-Vol sampler." Air Quality Laboratory Publication 3565/03/80/ARM, Texas A&M University, College Station, TX, 1980.
- (42) Wedding, J.B., Weigand, M., John, W. and Wall, S.: "Sampling effectiveness of the inlet to the dichotomous sampler." *Envir. Sci. Technol.* 14, 1367-1370- 1980.
- (43) Wedding, J.B.: "Ambient aerosol sampling: history, present thinking and proposed inlet for inhalable particulate matter." In *Proceedings: APCA Specialty Conference on the Technical Basis for a Size Specific Particulate Standard*, Air Pollution Control Assoc., Pittsburgh, PA, April 1980.
- (44) Shaw, R.W., Stevens, R.K. and Lewis, C.W.: "Comparison of aerosol inlets for dichotomous samplers." Submitted to *J. Air Poll. Contr. Assoc.*, 1981.

- (45) Coutts, H.J.: "A Study of Winter Air Pollutants at Fairbanks, Alaska." EPA/600/3-79-100, p. 21, Corvallis Environmental Research Laboratory, Corvallis, OR, September 1979.
- (46) Charell, P.R. and Hawley, R.E.: "Characteristics of water adsorption on air sampling filters." *Amer. Ind. Hyg. Assoc. J.* 42, 353-360, 1981.
- (47) Engelbrecht, D.R., Cahill, T.A. and Feeney, P.J.: "Electrostatic effects on gravimetric analysis of membrane filters." *J. Air Poll. Contr. Assoc.* 30, 391-392, 1980.
- (48) Jaklevic, J.M., Gatti, R.C., Goulding, F.S. and Loo, B.W.: "A beta-gauge method applied to aerosol samples." *Envir. Sci. Technol.* 15, 680-686, 1981.
- (49) Courtney, W.J., Shaw, R.W. and Dzubay, T.G.: "Precision and accuracy of a beta-gauge for aerosol mass determinations." Submitted to *Environ. Sci. Technol.*, 1981.



TLF. (02) 71 41 70

# NORSK INSTITUTT FOR LUFTFORSKNING

(NORGES TEKNISK-NATURVITENSKAPELIGE FORSKNINGSRÅD)  
 POSTBOKS 130, 2001 LILLESTRØM  
 ELVEGT. 52.

RAPPORTTYPE Oppdragsrapport	RAPPORTNR. OR 46/81	ISBN--82-7247-273-2
DATO NOVEMBER 1981	ANSV.SIGN. B.Ottar	ANT.SIDER 36
TITTEL A literature survey of methods for fractionated suspended particulate matter measurement.		PROSJEKTLEDER V.Vitols
FORFATTER(E) V. Vitols		NILU PROSJEKT NR 22181
		TILGJENGELIGHET **
OPPDRAAGSGIVER SFT		OPPDRAAGSGIVERS REF.
3 STIKKORD (å maks.20 anslag) Svevestøv	Størrelsesfraksjonering	Målemetode
REFERAT (maks. 300 anslag, 5-10 linjer)		
TITLE A literature survey of methods for fractionated suspended particulate matter measurement.		
ABSTRACT (max. 300 characters, 5-10 lines) Literature review suggests the US-type dichotomous sampler (virtual impactor) as the best commercially available device for suspended particle fractionation/gravimetric analysis at this time. The size selective high volume sampler lacks a reliable fractionator, but collects optimum particle mass for gravimetry. Field comparison trial of candidate samplers is recommended.		

\*\*Kategorier: Åpen - kan bestilles fra NILU           A  
 Må bestilles gjennom oppdragsgiver           B  
 Kan ikke utleveres                                   C