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A Field Intercomparison of Three Cascade Impactors

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ABSTRACT. Cascade impactors separate aerosol particles inertially and collect them for later analysis. While laboratory calibrations typically indicate performance close to design specifications, during field operation impactors are subject to a number of sampling artifacts, including particle bounce, inlet and internal losses, and particle size changes as pressure drops within the impactor.

To test the vulnerability of some commonly used impactors to these problems under field conditions, we participated in a shipboard intercomparison off the coast of Washington state between a micro-orifice uniform deposit impactor (MOUDI), a Berner low-pressure impactor, and a Sierra high-volume slotted impactor. Since there were some inconsistencies in the results, a second intercomparison was performed at Bellows Beach, Hawaii, between two MOUDIs and the Berner impactor.

Impactor samples were analyzed for soluble inorganic ions including Na^+, K^+, Cl^-, and NO_3^-, primarily from large (>1 μm) sea salt particles and NH_4^+, nonsea salt sulfate (NSS), and methanesulfonate (MS^-), found primarily in smaller aerosols.

The Sierra collected sea salt particles far more efficiently than the other impactors, which had severe inlet losses for 7 μm and larger particles. The MOUDI and Berner showed insignificant differences in the mass median diameter of accumulation mode particles (~0.34 μm), whereas the Sierra indicated almost twice the diameter (0.58 μm) of the others. AEROSOL SCIENCE AND TECHNOLOGY 29: 475–492 (1998) **© 1998 American Association for Aerosol Research**

INTRODUCTION

The aerosol size distribution in the marine boundary layer is usually multimodal with a supermicrometer (or "coarse") mode consisting primarily of sea salt and sometimes aeolian dust, and one or more submicrometer modes composed mainly of sulfates (Ahr and Pruppacher, 1989). The geochemical cycling and potential climatic importance of aerosol chemical species in the marine atmosphere depend strongly on how species are distributed with aerosol size. For exam-

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ple, supermicrometer aerosols are thought to account for most of the dry deposition of trace metals like iron and lead to the ocean surface (e.g., Arimoto et al., 1989). Both size and composition control aerosol response to humidity (Covert et al., 1990), which affects light scattering and cloud condensation nuclei concentrations.

Cascade impactors (CIs) collect ambient airborne aerosols and segregate them inertially into several aerodynamic size ranges for subsequent determination of chemical or physical properties. A variety of designs have appeared since May (1945) first reported use of a CI to sample coarse aerosols. Size cuts for currently available models range from about 10 μm to as small as 0.034 μm (Hillamo and Kauppinen, 1991; Hering et al., 1978). As such, they are well adapted to sample both accumulation mode (roughly 0.1 to 1 μm diameter) and coarse (>1 μm) particles. Because the samples are collected for later analysis, a wide variety of analytical techniques can be employed.

The fluid mechanics of impaction are well known, so laboratory calibrations often demonstrate performance close to design specifications (Marple et al., 1991; Hillamo and Kauppinen, 1991; Hering et al., 1979). However, field sampling conditions can cause significant deviations in collection characteristics. Particles can bounce from the collection surface or shatter and collect on later stages. This appears to be less of a problem for the liquid particles commonly found in the high relative humidities of the marine boundary layer than for solid particles (Stein et al., 1994). In windy conditions, inlets tend to discriminate against large particles, distorting recovered size distributions. Published algorithms based on wind tunnel data can be used to estimate these losses (Okazaki et al., 1987; Hangal and Willeke, 1990a), but it is unclear how reliable they are in the field.

As air is drawn through an impactor, its pressure drops. Water vapor pressure drops as well, thus lowering relative humidity, which can cause hygroscopic particles to shrink. However, aerodynamic cooling within the jets can raise relative humidity. These effects can cause substantial size changes, thus distorting the retrieved size distribution (Biswas et al., 1987). This is particularly severe with small particles at high ambient humidities and in impactors with large pressure drops.

TABLE 1. Operating Characteristics of the Cascade Impactors

	Sierra	Berner	MOUDI[a]
Flow rate[b]	1000	30	30
Substrates	Whatman 41	Tedlar	PTFE or Aluminum
Filters	Whatman 41	1 μm PTFE	1 μm PTFE
Size cuts[c]			18[d]
		8.0[d]	9.9
	7.0	4.0	6.2
	3.5	2.0	3.1
	1.4	1.0	1.8
	0.8	0.5	1.0
	0.4	0.25	0.56
		0.13	0.32
			0.18

[a] Each MOUDI is calibrated at the factory. Some of the stages differ by small amounts from the numbers here. Those differences are included in the inversions.
[b] Design flow rate, L m^{-1}.
[c] Nominal aerodynamic diameters of 50% size cuts, μm.
[d] Inlet stage (not usually analyzed).

Another practical difficulty with using cascade impactors is that the native data are not actually size distributions but an imperfect separation of particles into a few size bins. To reconstruct ambient size distributions from retrieved masses, it is necessary to invert the impactor data. This requires the use of calibrations to determine collection efficiencies of each stage as a function of particle size. Unfortunately, it is generally impossible to obtain a definitive solution, as the inversion problem is underdetermined and ill-posed (Crump and Seinfeld, 1982).

To examine the effects of these potential problems under practical conditions in the marine boundary layer, we performed two intercomparisons using three CIs of very different designs (Table 1). The Sierra impactor is a high-volume device with five slotted stages plus an after-filter. It has been used routinely in the marine boundary layer (e.g., Arimoto et al., 1985; Schneider et al., 1990). Willeke (1975) performed a careful calibration, including internal losses, interactions between stages, and the effects of different

impaction substrates. Unfortunately, he was limited to particles over a micrometer in size, so the last stage is not well characterized. Wedding et al. (1977) examined inlet losses for an enclosure similar to that commonly used with Sierra impactors. They found substantial losses with a 50% cutoff at about 15 μm in winds of 4.6 m s^{-1}.

The Berner low-pressure impactor (Berner and Lürzer, 1980) employs multiple circular jets in up to 10 impaction stages with nominal size cuts as small as 0.035 μm. The last stages collect those particles by operating at low pressure, thus reducing the amount of air available to deflect the particles away from the impaction surface. Several calibrations have been performed on various versions of the Berner impactor (Wang and John, 1988; Hillamo and Kauppinen, 1991). Due to the low-pressure design, humidity effects might distort size distributions. Wang and John (1988) concluded that size changes were negligible at up to 70% RH for stages with size cuts of 0.075 μm and larger, but humidities are often above that in the marine boundary layer. The same study reported low internal losses for this design.

The micro-orifice uniform deposit impactor (MOUDI) also uses circular jets and up to ten impaction stages plus an inlet stage. The last stage uses 2000 jets 52 μm in diameter to collect particles as small as 0.056 μm (Marple et al., 1991). The large number of very small jets reduces the pressure drop required to separate small particles, which should minimize size changes due to humidity effects. Alternating stages rotate to prevent accumulation of particle deposits under the jets. Complicated flow patterns within the impactor lead to fairly high internal losses for particles >6 μm. Tests for humidity effects within the MOUDI have shown significant distortions only at high humidities for the smallest available stages (Fang et al., 1991).

Kernel functions for the three impactors are shown in Fig. 1. They are derived from stage collection efficiency and internal loss data (Willeke, 1975 for the Sierra; Wang and John, 1988 and Hillamo and Kauppinen, 1991 for the Berner; and Marple et al., 1991 for the MOUDI). The MOUDI has the sharpest size cuts, minimizing cross-sensitivity between stages. In general, particles of any given size are collected on only one or two stages. The drops in efficiency in the large and small sizes are due to internal losses. The Berner is similar, though the curves are not quite so steep. In contrast, the Sierra kernel functions indicate large overlaps between stages.

EXPERIMENTAL

Sampling Procedures for the Shipboard Experiment

During the third Pacific Sulfur/Stratus Investigation (PSI-3) campaign in May 1991, we sampled near-surface marine aerosols aboard the NOAA ship R/V *Discoverer* from 50 to 250 km off the coast of Washington state. Patricia Quinn of the NOAA Pacific Marine Environmental Laboratory (PMEL) operated a six-stage low-pressure "Berner" impactor (Berner et al., 1979) manufactured at PMEL (there were actually seven stages, but the first, nominally 8 μm, was not analyzed). The NOAA Atlantic Oceanographic and Meteorological Laboratory (AOML) used a five-stage high volume slotted Sierra impactor from Sierra Instruments and very generously operated an eight-stage MOUDI (MSP Corporation) for the University of Rhode Island group. Operating characteristics of the three devices are summarized in Table 1.

All samplers were mounted on a railing just below and forward of the ship's pilot house at about 20 m above waterline and 30 m aft of the bow. The Berner impactor was mounted inlet down in a holder on the rail. It required no other protection. The MOUDI was mounted inside a sealed aluminum box about 15 cm square by 60 cm high, which also contained the rotator motor and pressure gauges. The impactor inlet protruded through the top of the box. To reduce rainfall into the MOUDI without making a convoluted flow path, a 23 cm square plastic tile was mounted about 15 cm above the

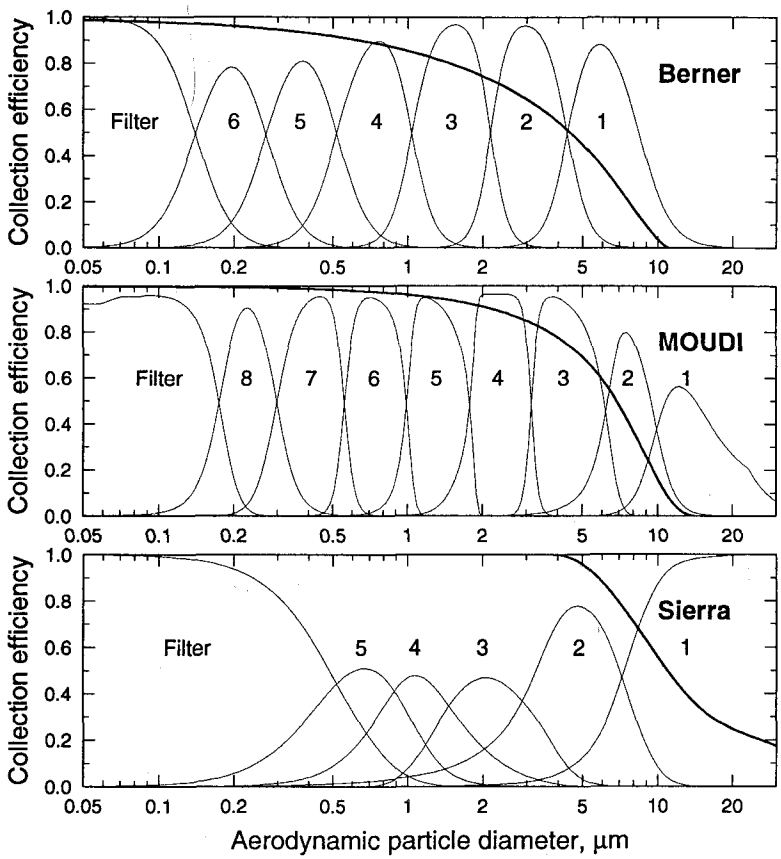

FIGURE 1. Kernel functions for the three impactors. The curves were generated from published calibrations. The heavy lines are estimated inlet efficiencies for a wind of 5 m s^{-1}. Berner and MOUDI losses are calculated from Hangal and Willeke (1990b) whereas Sierra losses are from Wedding et al. (1977).

inlet. The Sierra impactor was mounted in a rigid polyethylene shed, requiring air to flow up around the sampler before reaching the inlet.

To reduce the possibility of contamination from ship exhaust, a sector controller restricted sampling to periods when the relative wind was greater than one meter per second and ahead of the beam. Sampling periods were identical for all of the impactors. There were ten sampling periods during the cruise, ranging from 15 to 28 hours. The MOUDI rotator motor failed during the last interval, so no sample was available. Period 10 was therefore excluded from all calculations, except for the NH_4^+ comparison, which involved only the Sierra and Berner impactors.

Berner impactor procedures. Prior to use aboard ship, the Berner impactor was rinsed with distilled, deionized water, and then with spectral grade methanol. Tedlar films were used as impaction substrates in the impaction stages, and 47 mm diameter Millipore Fluoropore 1 μm pore size polytetrafluoroethylene (PTFE) filters were used to collect particles that penetrated all of the impaction stages. Before being loaded into the impactor, Tedlar films were agitated in an ultrasonic bath in 10% H_2O_2 for one hour, then rinsed six times with distilled, deionized wa-

ter. They were dried in an NH_3- and SO_2-free glove box.

During sampling, airflow through the impactor was maintained at 30 sLpm (defined at 20°C, 1.013×10^5 Pa) by a mass flow sensor (Hastings HFM-C201) coupled with a regulating valve.

Immediately after sampling, the filter and impaction films were put into separate polyethylene test tubes and wetted with 1 mL of spectral grade methanol followed by 5 mL of distilled, deionized water. They were ultrasonically agitated for 30 minutes and poured into polyethylene scintillation vials. All analyses were done by ion chromatography on board the ship immediately after collection as described by Quinn et al. (1993). Analytical uncertainties were ±1.5% for NO_3^-, $SO_4^=$, Na^+, and NH_4^+ and 3% for MS^- (95% confidence limits). Flow uncertainties were estimated to be ±10%, yielding overall uncertainties of 11% to 13%.

Sierra impactor procedures. Prior to each sampling interval all parts of the Sierra impactor were scrubbed with nonionic detergent solution, rinsed with deionized water, and air dried in a class 100 clean bench in the ship's laboratory. Pre-cut Whatman 41 impaction substrates and 20 × 25 cm Whatman 41 final filters were used as received from the vendor.

Airflow rate was monitored with a sharp-edged orifice flow tube that was calibrated in situ with a static pressure calibrator (General Metals model GMW-25) prior to the cruise. Airflow rate was maintained at 1.1 ± 0.1 $m^3\ min^{-1}$ by manual adjustment of pump speed with a Variac (checked hourly).

At AOML after the cruise, two central strips were cut from each impaction substrate, and two 47 mm diameter disks were punched from each final filter. These samples were extracted in 10 mL of deionized water, sonicated for 20 minutes, filtered through Gelman AcroDiscs, spiked with 20 μL of chloroform to inhibit biological activity, and stored at room temperature in polyethylene bottles until analysis.

Analyses were done using automated ion chromatography at AOML. Estimated analytical relative standard errors were ±9% for Cl^-, NO_3^-, SO_4^{-2}, and Na^+, ±12% for MS^-, and ±15% for NH_4^+. Estimated flow rate uncertainty was ±12%. These uncertainties combine to yield overall uncertainties for individual species of 15% to 19%. Examination of the initial results revealed an interference in chromatograms that caused underestimations of MS^- peak areas by factors of 2–3. To remove this interference all extracts were reanalyzed using a linear gradient in eluent concentration.

MOUDI impactor procedures. All components of the MOUDI were cleaned at URI prior to the cruise. A stainless steel punch was used to make 47 mm diameter impaction substrates from 25 μm thick PTFE film. These were used for the eight impaction stages and the inlet stage, whereas the backup filters were PTFE with 1 μm pore size (Fluoropore filters from Millipore). Immediately after sampling, substrates and the filter were removed from the impactor and heat sealed in separate microclean polyethylene bags (Clean Room Products). Samples were stored at room temperature until analysis after the cruise at URI.

A hot wire mass flow controller (Tylan model FC262V) was used to maintain constant airflow through the MOUDI. Calibration after the cruise revealed that the flow was about 10% below the intended 30 sLpm, which increased cut sizes by 5.4% (Howell, 1996).

At URI, impaction films and filters were extracted with 10^{-5} M dichloroacetic acid. Filters were wetted with ethanol first. All were agitated ultrasonically for 15 minutes. Analyses were done by ion chromatography. As a result of contamination from glass autosampler vials, Na^+ was reanalyzed by flame atomic absorption spectroscopy. Analytical uncertainties were ±3% for SO_4^{-2} and ±5% for other species. Flow rate uncertainties were estimated at ±5%. These combine to make overall uncertainties for individual species of 6% to 7%.

Procedures at Bellows Beach

As shown in Tables 2–4, the shipboard intercomparison revealed major discrepancies,

so another intercomparison was performed in the summer of 1993 on a 20 m tower on the beach of Bellows Air Force Station, Oahu, Hawaii. The Sierra impactor was not available, but a second MOUDI with slightly different characteristics and a different enclosure was used. The Berner impactor was used for 5 of the 11 sampling periods, which ranged from 3 to 101 hours. Due to the strong prevailing trade winds and the windward location of the beach, no sector controller was used. Waves breaking on an offshore reef may have enhanced concentrations of sea salt particles.

The instruments were mounted on or near the top rail at the eastern corner of the sampling platform at about 20 m altitude and 25 m from the shore. Each impactor was used in a variety of mounting configurations to explore of the effect of airflow around the samplers. The MOUDIs were mounted either in the enclosure used in PSI-3, an unsealed enclosure supplied by MSP, or were tied down to a plywood sheet atop the tower without a rotator mechanism. The Berner was generally mounted as in PSI-3, but was once mounted horizontally on the plywood sheet.

The PSI-3 MOUDI enclosure was modified by the addition of aluminum L-shaped struts on each corner to support the rain shield, which had been supported by a 5 cm tube about 12 cm behind the impactor on the ship.

The Berner impaction substrates were Tedlar film, as in PSI-3. Zefluor PTFE filters (Gelman) were used as after-filters. Aluminum foil substrates were used in the MOUDIs, except for the last two samples, when PTFE films were also used. We chose Al substrates because they do not tend to lift the substrate hold-down rings and distort airflow through the stages. Foils were used as received from MSP.

Mass flow controllers maintained constant flow through the MOUDIs. As during PSI-3, flow was about 10% below 30 sLpm. Flow through the Berner was adjusted with a needle valve and monitored with a TSI 2014L-V hot wire flow sensor. Its error was estimated at ±8%. Samples were sealed individually in clean polyethylene bags and stored at room temperature until analysis.

Analyses were limited to Na^+ and SO_4^{-2}, taken to represent sea salt and accumulation mode particles. All impaction substrates were extracted with 10^{-5} M oxalic acid. Filters were wetted with 1 mL of ethanol first. Ultrasound was not used for impaction substrates because it caused the aluminum foil to crumble. Ion chromatographs were used to analyze the extract. Estimated analytical errors were 6% for SO_4^{-2} and 10% for Na^+.

Inversions

Two inversion techniques are used here. The simplest and perhaps most commonly used method is to ignore all calibration data except for the particle diameter at which each stage captures 50% of the incident aerosols (D_{50}) and to assume that each stage captures all particles between its D_{50} and that of the previous stage. The aerosol size distribution within that range is assumed to be constant. The result looks like a histogram (Fig. 2). This method has the advantage of simply and unambiguously representing the raw data. The fact that the resulting size distributions are clearly not the same as that of the ambient aerosol is ameliorated somewhat by the impossibility of proving that any other method produces correct answers. While useful for visual presentations, the histogram inversion is inappropriate for comparisons of CIs with different size cuts, numbers of stages, and internal loss mechanisms.

It is preferable to use an inversion that uses all available calibration data and produces results that can be compared easily. The technique used here, adapted from Dzubay and Hasan (1990), assumes that ambient distributions consist of one or two lognormal modes. A nonlinear least squares fit is then performed to determine median diameter, total mass, and width (geometric standard deviation) of each mode (Fig. 3). This is a relatively straightforward procedure that produces parameters that are easy to compare. The primary drawback of this method is that the real distributions may not

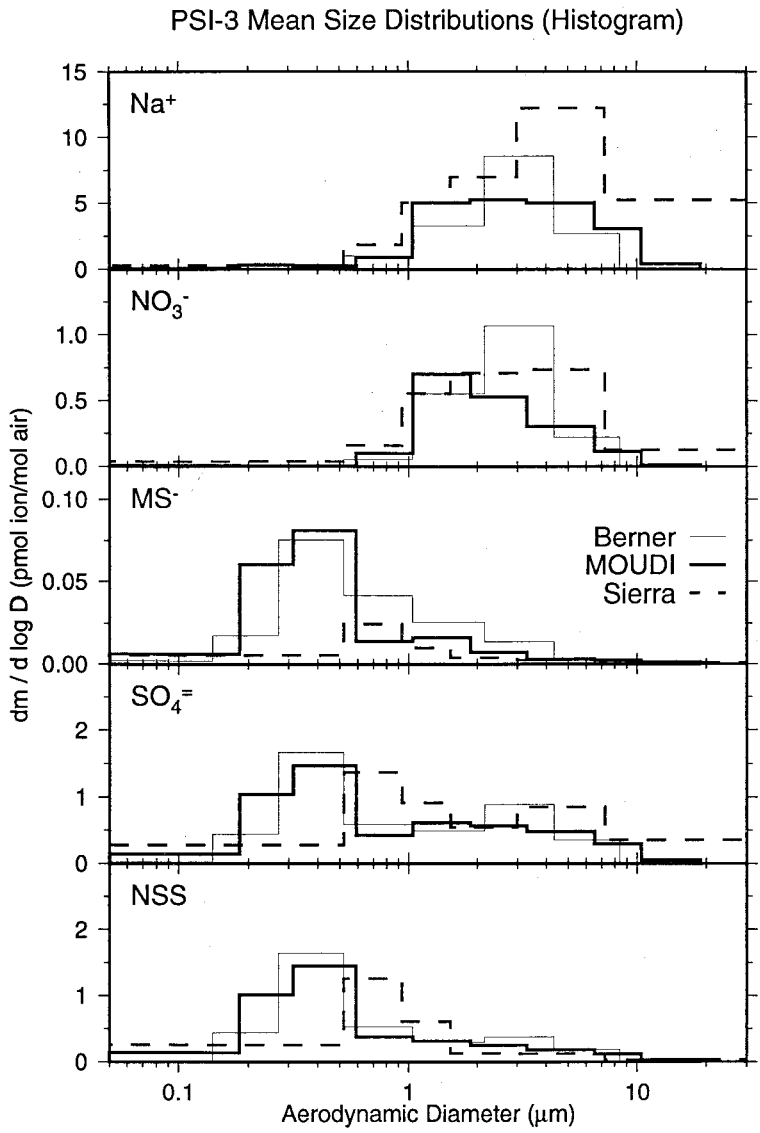

FIGURE 2. PSI-3 project average size distributions. Masses on each stage are divided by the logarithm of the ratio of D_{50} of that stage and the previous one.

be lognormal. In addition, at least three good data points are required to fully define each lognormal mode. As the Sierra has only five impaction stages and the Berner we used had just six, modes spanning just two stages occasionally occurred, rendering accurate bimodal fits impossible. Nevertheless, lognormal fitting appears to produce consistent mass median diameters even in the presence of analytical errors (Howell, 1996), so we have used it to compare the three impactors.

Error Calculation Procedures

There are two fundamental types of errors associated with ion chromatography, used by

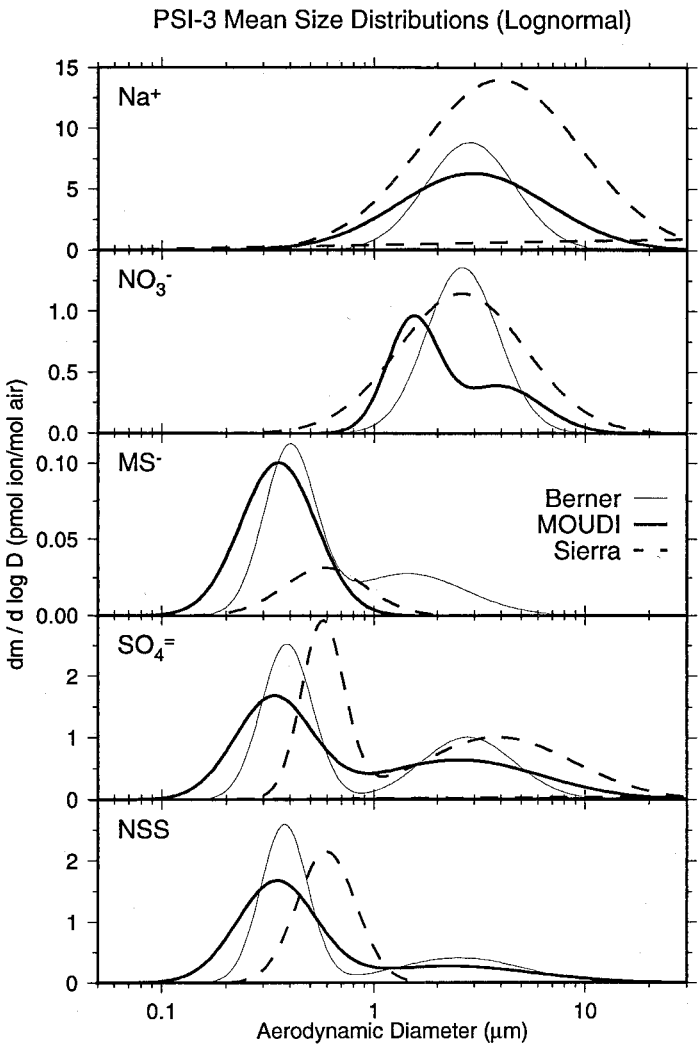

FIGURE 3. PSI-3 project average size distributions inverted using a least squares fit to lognormal modes (Dzubay and Hasan, 1990). Bimodal fits were attempted in each case. When no convergence was achieved, a unimodal fit was used.

each group for sample analysis. Changes in cell sensitivity and injection volume give errors proportional to concentration. Estimates of these errors from each laboratory are given below. In addition, baseline noise, which determines the detection limits, adds an error independent of concentration. Detection limits were different for each laboratory and each species, but were generally a few hundred nanomoles per impactor stage. Because none of the species examined here is uniformly distributed across all sizes, there are usually one or more stages with concentrations near or below detection limits. Therefore, it is vital to include the fixed errors when performing inversions to prevent attempts to fit small (or zero) concentrations very closely. It is particularly important when fitting lognormal curves, which by definition cannot have zero concentrations in any size range.

In contrast, the proportional errors generally dominate the overall concentration, as there are usually a few stages well in excess of detection limits. Those must be combined with flow measurement inaccuracy to determine an overall error estimate.

TABLE 2. Total Concentrations (Sum of All Stages) During PSI-3. B Refers to the Berner Impactor, M to the MOUDI, and S to the Sierra

	Sample Interval[a]									
	1	2	3	5	6	7	8	9	10[b]	Ratio[c]
					Na^+, nmol m^{-3}					
B	45	34	36	10	19	43	44	89	115	0.64 ± 0.16
M	42	31	46	23	27	27	54	122	—	0.75 ± 0.15
S	67	59	71	43	76	114	131	244	137	1.60 ± 0.21
					NH_4^+, nmol m^{-3}					
B	7.8	3.2	4.4	6.3	2.4	2.0	6.2	7.9	8.9	1.23 ± 0.10
S	6.6	2.4	3.5	3.8	1.4	1.0	3.4	4.9	4.2	0.77 ± 0.10
					MS^-, nmol m^{-3}					
B	0.21	0.14	1.04	0.86	0.20	0.31	0.38	0.15	0.77	1.30 ± 0.26
M	0.34	0.23	0.99	0.70	0.20	0.22	0.21	0.20	—	1.33 ± 0.33
S	0.06	0.10	0.43	0.09	0.06	0.11	0.10	0.00	0.04	0.36 ± 0.37
					NO_3^-, nmol m^{-3}					
B	10.	2.9	3.9	3.4	2.1	2.8	5.9	5.7	5.9	0.97 ± 0.17
M	10.	3.9	1.1	2.2	1.2	1.6	6.3	3.2	—	0.69 ± 0.24
S	12.	4.2	4.3	7.8	3.0	3.3	8.2	6.9	24.	1.33 ± 0.20
					NSS-Sulfate, nmol m^{-3}					
B	11.0	6.6	10.8	12.2	4.4	4.8	6.7	9.3	13.0	1.12 ± 0.18
M	14.6	6.7	10.4	11.8	3.1	2.6	5.1	13.2	—	1.03 ± 0.19
S	11.8	4.6	7.9	9.1	2.8	2.4	6.8	7.0	8.9	0.84 ± 0.12

[a] Interval 4 was a blank (impactors mounted but no airflow) so is not included.
[b] There was no MOUDI sample during interval 10. Data for this interval were used only for the NH_4^+ comparison.
[c] Average and standard deviation of the ratio of sampler value to average of sampler values from each interval.

Flow errors are less important in determining size distributions, because each stage is affected equally. Impaction is governed by the Stokes number, which varies as the square root of the flow rate.

Complete procedural blanks were performed for each impactor during PSI-3 and at Bellows. Impactors were prepared as usual, but no air was drawn through them. Blank values were subtracted from each sample, though this correction was inconsequential as blank values were generally below detection limits.

RESULTS

Laboratory Intercomparisons

Stock solutions from which each lab made calibration standards were compared at AOML. Mean measured concentrations for each laboratory's standards agreed to within 7% or better of the overall mean, and six of eight were within 3%. In addition, each lab measured Na^+, NO_3^-, and SO_4^{-2} in two NIST artificial rainwater standard solutions. Means for each lab agreed to within 7% of the certified values, though the certifications were void, because several months had passed since the expiration dates. In summary, differences between stock solutions were small and can account for only a minor fraction of the discrepancies observed during the intercomparison.

PSI-3

Total concentrations (the sum of all stages) for each species measured during the PSI-3 project are listed in Table 2. A two-factor analysis of variance (Zar, 1974) was used to determine whether significant differences exist at the 95% confidence level. This was followed by a Newman-Keuls multiple range test to determine which pairs of samplers differed.

Significant differences were present in each species. For NO_3^- and Na^+, the Sierra masses were higher than the Berner and MOUDI, which were indistinguishable. These species were found primarily on

TABLE 3. Inversion Results for Na^+ from PSI-3

	Sampling Period[a]									
	1	2	3	5	6	7	8	9	10	Ratio[b]
Mass in mode (nmol m^{-3})										
B	46	35	34	—	20	42	44	86	113	0.67 ± 0.17
M	42	32	42	22	23	—	—	114	—	0.72 ± 0.10
S	72	63	77	48	85	127	145	275	152	1.60 ± 0.26
Median aerodynamic diameter (μm)										
B	3.4	3.5	2.7	—	3.3	2.7	3.1	2.5	3.0	0.82 ± 0.07
M	3.5	3.7	2.8	4.1	5.4	—	—	2.6	—	0.92 ± 0.13
S	4.8	4.7	4.8	3.7	5.5	3.9	3.7	4.0	3.6	1.26 ± 0.09

[a] The period 5 Berner data and the periods 7 and 8 MOUDI data were multimodal so cannot be directly compared with the other samplers.
[b] Average ratio of sampler value to mean of all samplers (only periods 1, 2, 3, 6 and 9).

larger size particles, as is generally the case in the marine boundary layer (Savoie and Prospero, 1982). The quantity of Na^+ retrieved by the Sierra was generally more than twice that of the other impactors. In contrast, the Sierra found less MS^- than the other impactors. NSS from the Sierra was lower than that from the Berner. The MOUDI NSS was between the others, but not significantly different from either.

When the data were inverted, the Sierra indicated a larger median particle size for Na^+ than found by the other impactors (Table 3). The coarse mode peak generally appeared to be about 4.4 μm, whereas the other two indicated the peak was about 3.2 μm.

Figures 2 and 3 show average size distributions for the entire project. The lognormal inversions (Fig. 3) enable a more direct comparison but lack the detail present in the histograms. This is particularly severe for the Sierra impactor, whose five stages plus filter provide insufficient information for a bimodal fit. Consequently, the minor mode often fails to converge, tending toward an extremely thin, high peak. These were eliminated from the figure.

While NSS accumulation mode mass varied considerably among both sampling periods and impactors, there were no statistically significant differences among samplers (Table 4). In contrast, the Sierra reported distinctly larger mass median diameters (averaging 0.58 μm) than the other two impactors. The MOUDI and Berner averaged 0.34 and 0.37 μm respectively, but the difference was not statistically significant. The smaller numbers correspond better to the 0.3 μm mass peak typical for marine accumulation mode aerosols (Whitby, 1978).

A peculiar feature of the NSS data is that the Sierra tends to have a smaller fraction of its NSS mass in the coarse mode. A histo-

TABLE 4. Inversion Results for Accumulation Mode NSS from PSI-3

	Sampling Period[a]									
	1	2	3	5	6	7	8	9	10	Ratio[b]
Mass in mode (nmol m^{-3})										
B	11.7	5.4	8.5	10.2	3.2	2.4	3.3	2.6	11.3	1.01 ± 0.25
M	10.0	4.8	7.0	10.4	2.3	—	—	6.8	—	1.01 ± 0.16
S	10.9	4.3	7.6	8.5	2.5	2.1	6.1	6.3	8.6	0.98 ± 0.12
Median aerodynamic diameter (μm)										
B	0.35	0.38	0.49	0.37	0.33	0.33	0.37	0.34	0.38	0.87 ± 0.06
M	0.38	0.31	0.39	0.34	0.27	—	—	0.35	—	0.78 ± 0.05
S	0.61	0.58	0.72	0.55	0.47	0.56	0.57	0.58	0.58	1.35 ± 0.03

[a] The periods 7 and 8 MOUDI were not successfully inverted.
[b] Average ratio of sampler value to mean of all samplers (excluding periods 7, 8, and 10).

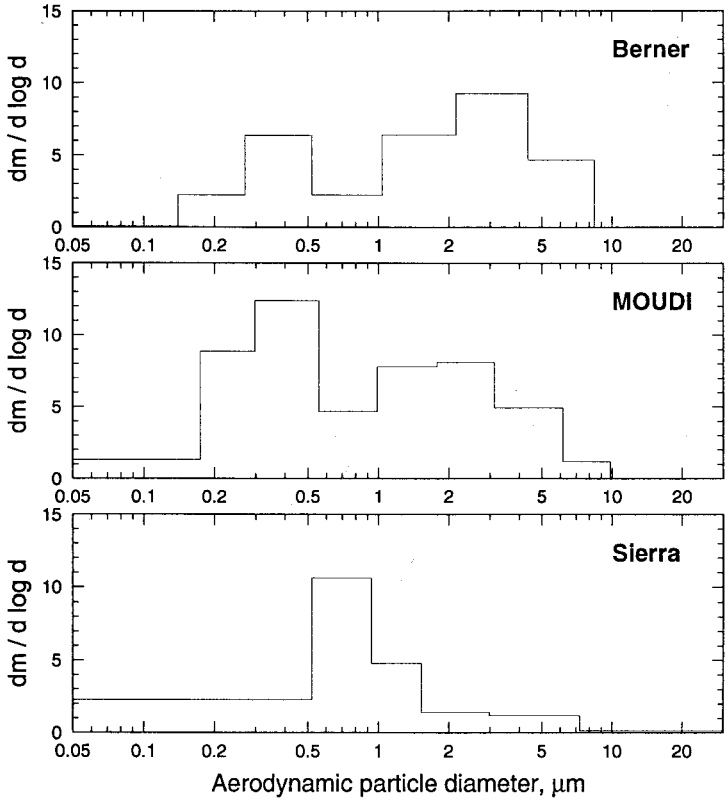

FIGURE 4. Histogram inversions of NSS data from sampling period 9 of PSI-3.

gram inversion of the most extreme example is shown in Fig. 4. This is the main reason that the inversions show accumulation mode mass to be indistinguishable among samplers while the total NSS collected by the Sierra is significantly smaller (Table 2). Another contributing factor is that because mass median diameter of the accumulation mode is larger, the tail extends to include larger diameters. In addition, the inversion process compensates for internal losses of the Sierra, though those losses are only about 3% for 1 μm particles, so this is a minor correction.

Bellows

The Bellows data (Table 5) indicate that the quantity of Na^+ found is extremely sensitive to airflow around the sampler and its mounting. When mounted identically, as in periods 8, 10, and 11, the two MOUDIs generally gave similar results. In the other samples, when they were mounted differently, the two MOUDIs differed dramatically. Data from period 7 are shown as an example in Fig. 5. The ship enclosure clearly enhanced sampling efficiency compared with the enclosure supplied by MSP. This enhancement starts below 2 μm diameter and increases with diameter. Tests with a handheld hot wire anemometer indicated that the mounting struts for the rain shield of the ship enclosure slowed airflow considerably. In contrast, there were no struts upstream of the MSP enclosure, and airflow accelerated past the inlet. This acceleration also affected the MOUDIs when not installed in the enclosures.

During sampling period 9, MOUDI 2 and the Berner lay horizontal facing into the wind while MOUDI 1 stood upright. Inlets were not truly isoaxial because the wind on

TABLE 5. Total Masses (Sum of All Stages) of Na^+ and NSS Collected during the Bellows Intercomparison. Inlet Stages of the MOUDIs are not included.

	Sampling Period										
	1	2	3	4	5	6	7	8	9	10	11
	NSS (nmol m^{-3})										
M1	2.6	3.6	4.3	2.1	2.7	6.2	4.8	4.8	5.3	3.4	2.2
M2	2.7	3.5	5.4	2.4	2.9	5.8	5.1	5.1	5.1	3.3	3.8
B	—	—	—	—	—	—	6.9	5.8	6.6	3.8	4.1
	Sampler mounting and orientation										
M1	[⊓]	[⊓]	⟨⊓⟩	[⊓]	[⊓]	[⊓]	⟨⊓⟩	⊓	⊓	⊓	⊓
M2	⟨⊓⟩	⟨⊓⟩	[⊓]	⟨⊓⟩	⟨⊓⟩	⟨⊓⟩	[⊓]	⊓	▭	⊓	⊓
B	—	—	—	—	—	—	⊓	⊓	⊓	⊓	⊓
	Na (nmol m^{-3})										
M1	147	118	58	89	174	86	60	84	192	71	75
M2	58	57	111	48	64	46	178	95	267	75	129
B	—	—	—	—	—	—	68	70	202	59	73

Key to sampler mounting:

- ⊓ an unenclosed MOUDI in the normal (inlet up) position.
- [⊓] a MOUDI in the PSI-3 sealed ship enclosure with rain shield (see text).
- ⟨⊓⟩ a MOUDI in the standard MSP enclosure.
- ⊓ teflon substrates used instead of aluminum.
- ▭ an unenclosed MOUDI mounted horizontally, inlet into the wind.
- ⊓ Berner impactor mounted normally (inlet down).
- ⊓ Berner mounted horizontally, facing the wind.

the tower was directed upward by the shoreline and the trees on it. The effect was similar to that of the ship enclosure, with larger amounts collected by the horizontal MOUDI. With the exception of the inlet stage, which indicated 430 nmol m^{-3} Na^+, the increased efficiency was somewhat less than that due to the ship enclosure. It is not clear whether the Berner collection efficiency increased.

In general, the Berner Na^+ results agreed better with the MOUDIs when the latter were not mounted horizontally or in the ship enclosure. Because the Berner and MOUDI 1 Na^+ did not differ much during the PSI-3 project, they must have had similar inlet losses. This suggests that the MOUDI inlet losses during PSI-3 must have been closer to the losses in the MSP enclosure than to the losses in the ship enclosure modified with the rain shield struts.

While sensitivity to Na^+ changed dramatically, the NSS totals appeared unaffected by mounting details and showed relatively good agreement between the two MOUDIs. However, the Berner consistently collected more NSS than the MOUDIs, by an average of 27%.

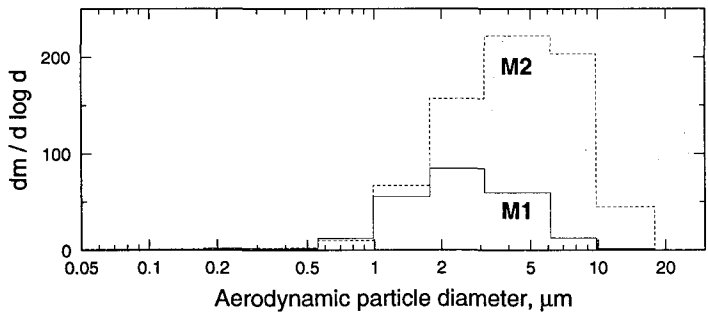

FIGURE 5. Effect of enclosures upon Na^+ collection in MOUDIs. Data are from Bellows sampling period 7. M2 was in the ship enclosure used in PSI-3, M1 in that supplied by MSP. The disparity appears to be due to flow speed past the inlet.

The discrepancy in NSS is primarily due to the accumulation mode, as is evident in the inverted data (Table 6). Mass in the accumulation mode peak was about 40% higher in the Berner. In contrast, the median diameters of the peaks are indistinguishable.

DISCUSSION

Sierra versus Berner and MOUDI

None of the samplers can be expected to gather large particles efficiently in the wind conditions experienced during PSI-3. Wedding et al. (1977) determined that inlet efficiencies for an enclosure similar to the one used for the Sierra dropped to 50% for 15 μm particles at wind speeds of 5 m s^{-1}, but they did not report sensitivity to wind speed. The average wind speeds recorded by the anemometer during PSI-3 was about 7 m s^{-1}. The wind speeds experienced by the samplers are unknown, since they were not located near the anemometer and airflow must have been dramatically altered by the presence of the ship. No comparable studies have been performed for the Berner or MOUDI and their enclosures, but algorithms for calculating inlet efficiencies of thin-walled tubular inlets are available (Hangal and Willeke 1990a). At 5 m s^{-1}, inlet losses should be at least 50% for 7 μm particles. Much better Sierra collection efficiencies above 7 μm would satisfactorily explain both the higher masses collected and the larger median particle diameters indicated by the Sierra impactor for Na^+. The NO_3^- totals point to the same conclusion.

The Bellows data emphasize the severity of the inlet problem. The MOUDI rain shield supports slowed the wind speed

TABLE 6. Inversion Results for Accumulation Mode NSS from Bellows

	Sampling Period											
	1	2	3	4	5	6	7	8	9	10	11	Ratio[a]
Mass in mode (ng m^{-3})												
M1	1.88	2.74	2.63	1.55	2.19	4.80	4.16	3.68	3.15	2.46	1.38	1.00 ± 0.18
M2	1.97	2.63	2.74	1.87	2.30	3.93	4.10	3.79	2.36	1.72	2.50	1.00 ± 0.18
B	—	—	—	—	—	—	5.32	4.86	5.08	2.62	2.72	1.41 ± 0.24
Median aerodynamic diameter (μm)												
M1	0.30	0.30	0.35	0.34	0.30	0.32	0.31	0.32	0.31	0.30	0.30	1.01 ± 0.03
M2	0.31	0.30	0.31	0.31	0.31	0.33	0.29	0.30	0.30	0.30	0.30	0.99 ± 0.03
B	—	—	—	—	—	—	0.29	0.32	0.33	0.29	0.30	1.01 ± 0.05

[a] Average ratio of sampler value to mean of the two MOUDI samplers. Calculated for periods 7–11 only.

enough that the amount of Na^+ collected was essentially doubled. It is tempting to use inlet loss calculations to correct for this during the inversion. However, because the losses are sensitive to alteration of local airflow due to small changes in mounting configurations, it is unclear how realistic such calculations can be (Howell, 1996). A better solution would be a high-efficiency inlet system. A shrouded sampler like that of McFarland et al. (1989) might be appropriate if wind direction were relatively constant or an omnidirectional inlet such recommended by Liu and Pui (1981) could be appropriate for use on a tower (though the efficiency of the latter was tested only for wind speeds up to 2.5 m/s).

The smaller retrieved masses of NH_4^+, MS^- and nonsea salt SO_4^{-2} in the Sierra results are more difficult to explain. All are typically found on accumulation mode particles, suggesting that somehow the Sierra collects fewer small particles than the others. It is conceivable that some flow measurements were inaccurate, but errors of 25% seem unlikely.

All three ions or their photochemical precursors are present in the gas phase in the marine boundary layer, so deposition to or evaporation from the impaction substrates could occur. This is particularly true of ammonia, which has a significant vapor pressure (Quinn et al., 1992). NSS is nonvolatile at ambient temperatures, but conversion of SO_2 to $SO_4^=$ is possible. However, this potential artifact appears unimportant on PTFE filters (Appel et al., 1984) so seems improbable on the MOUDI and Berner substrates. Such artifacts have been reported for the Whatman 41 substrates used in the Sierra (Appel et al., 1984), though appear unimportant in clean marine situations (Pszenny et al., 1993). In any case, artifact $SO_4^=$ production in the Sierra cannot explain the lower mass found by that device.

The use of Whatman 41 filters for aerosol sampling has been a source of controversy, as some studies have shown that they collect submicrometer particles inefficiently—missing as much as 85% of smoke particles and 56% of 0.1 μm particles (Harrison, 1987). This inefficiency appears to be somewhat inconsistent, but depends on face velocity and filter loading. However, typical losses appear to be <10% for bulk high volume sampling (Lowenthal and Rahn, 1987) and negligible when used in cascade impactors in the marine boundary layer (Pszenny et al., 1993).

Another possibility is that internal losses for submicrometer particles may be high. Willeke (1975) found internal losses of up to 14% for 3 μm particles, but was unable to test aerosols much below 1 μm diameter. The primary cause of internal losses of coarse particles is inertial impaction on the sides of the jets. This mechanism is less effective for small particles, so just 1% of 0.7 μm particles are lost. Very small particles can be lost be diffusion to internal surfaces, though in other impactors, such as the MOUDI, this is significant only at diameters below 0.1 μm. Unless diffusion of submicrometer particles to coarse mode impaction substrates is significant, it is unlikely that internal losses of accumulation mode particles in the Sierra is a major problem.

Because airflow through the impactor may not be uniform, some areas of the impaction substrates may gather more or less material than other areas. Because only central strips of the Sierra samples were analyzed rather than the entire substrates, it is conceivable that uneven deposition could account for part of the NSS shortage.

We have no good explanation for the very small concentration of MS^- found by the Sierra impactor. There is no obvious way to aerodynamically separate MS^- containing particles from others, so MS^- must be collected as efficiently as NSS, of which the Sierra collects ~80% as much as the other impactors (Table 2). Saltzman et al. (1983, 1986) demonstrated that MS^- on filters and in filter extracts is unaffected by storage at 5°C for periods of up to three years. As the Sierra samples were stored at room temperature, chemical destruction is conceivable, though the chloroform should have eliminated any biological losses.

The differences in NSS accumulation mode median diameters from the inversion

results pose another problem. Part of the answer might be that the Sierra has only one submicrometer stage and the after-filter available to capture accumulation mode particles, so its size resolving capability is too limited. Heintzenberg et al. (1981) state that meaningful inversions at a given particle size are possible only if the ratio of some two kernel functions changes as a function of particle size. An examination of the kernel functions (Fig. 1) suggests that perhaps the Sierra inversion ought to resolve particles as small as 0.2 μm, below the mass median diameters reported by the other impactors. We tested this by using the Sierra kernel functions and a simulated lognormal aerosol size distribution with a mass median diameter or 0.3 μm and finding the corresponding stage masses M_i. When those results were inverted the initial distribution was accurately reproduced, indicating that the Sierra should be able to resolve accumulation mode peaks, at least under ideal circumstances with a single mode, no analytical errors, and perfectly characterized kernel functions.

A more plausible explanation for the differences in NSS median diameter is inaccurate kernel functions due to inaccurate calibrations or inadequately characterized losses. Willeke (1975) was essentially unable to calibrate the last stage of the Sierra, since he could not generate particles smaller than a micrometer. Therefore, the theoretical 50% size cut was used to generate the kernel functions. However, Willeke showed that size cuts in the Sierra were quite sensitive to the presence of earlier stages and to the nature of the impaction surface. Stage collection efficiencies near theoretical values were obtained only with smooth impaction surfaces. The manufacturer recommends use of a thick fiberglass filter material to prevent particle bounce (Sierra Instruments Inc., 1971), but this dramatically increases collection efficiency for small particles in the largest few stages, apparently due to flow penetrating the filter material and allowing deposition by impaction on and diffusion to the fibers. It is reasonable to expect the same on the final stages. No calibrations have been performed with the Whatman 41 impaction substrate used during PSI-3. It is a fibrous material like the Gelman type A filter material tested by Willeke, but it is much thinner. The data gathered during PSI-3 suggest that the last stage of the Sierra impactor may gather many more small particles than the theoretical kernel function used here would suggest. If enhanced collection of fine particles on the last stage are not reflected in the kernel functions, median diameters returned by inversions would be too high.

Similarly, if there are actually major losses of submicrometer particles as discussed above, inversion results would be skewed to larger sizes.

Berner versus MOUDI

Though the MOUDI and Berner often showed large differences for individual sampling periods during PSI-3, there were no statistically significant overall patterns. During the Bellows intercomparison, disparities in the Na^+ results can be attributed to inlet losses. However the Bellows NSS data are difficult to interpret. The mass median diameters were always within 0.03 μm, but the mass collected differed sharply, with the Berner indicating about 40% more in the accumulation mode and roughly 25% more overall. As mentioned above, artifact SO_4^{-2} production on the Tedlar substrates is unlikely. Poor extraction efficiency from the aluminum substrates might be a possible explanation. The tests comparing PTFE and aluminum substrates were performed to examine that possibility. Though they did not show convincing evidence that Al performs much worse than PTFE, there were only two of these tests, with inconsistent results, so the possibility cannot be completely discounted.

The very close agreement in mass median diameter in both projects is a bit surprising, since we expected that humidity changes inside the impactors could significantly change particle diameters. These effects were explored in the MOUDI by Fang et al. (1991). Using a version of the impactor with smaller size cuts, they showed that the size cuts for

sulfuric acid particles increased by about 5% at 80% RH in a stage with a size cut of 0.122 μm and increased rapidly at higher humidities. This is primarily due to growth of the particles due to aerodynamic cooling within the jets that raises relative humidity. Smaller effects might be expected in the MOUDI used here, since the smallest size cut was 0.18 μm, and average humidity exceeded 80% only during the third PSI-3 sampling period and Bellows periods 4 and 11.

While the outlet pressure from the MOUDI is about 0.7 atm, the exit from the final stage of the Berner is about 0.2 atm, so relative humidity effects might well be larger. Particle shrinkage due to reduced absolute humidity would result, shifting the Berner peak farther downward than the MOUDI. However, it appears that particle growth due to aerodynamic cooling is a more important effect in low pressure impactors (Biswas et al., 1987). Wang and John (1988) used Biswas's calculations to predict a 50% growth in 0.168 μm $(NH_4^+)_2SO_4^{-2}$ particles at 70% RH. They rejected that conclusion when they failed to observe such large changes during controlled humidity tests. They concluded that residence times in the jets were too small for that much growth to occur. However, inverting their Figure 7 reveals that mass median diameter increased from 0.35 to 0.41 μm as the humidity increased to 69%. Since the humidity during both intercomparisons generally ranged between 70% and 80%, size increases of more than 0.06 μm appear reasonable. The lack of such results may indicate that ambient particles are less hygroscopic than $(NH_4^+)_2SO_4^{-2}$ or that humidity effects in the Berner are indeed small and the apparent changes in Fig. 7 of Wang and John (1988) are not a product of humidity differences.

CONCLUSIONS

The Sierra impactor appears to collect supermicrometer particles much more efficiently than do the Berner impactor or the MOUDI when configured as in PSI-3. As such, it is likely to give a better view of sea salt particle size distributions.

Poor inlet efficiency in the Berner and MOUDI is due to tubular inlets perpendicular to the wind. If reliable results are to be obtained for particles larger than a few micrometers, the standard inlets must be modified. Simply slowing airflow with upstream spoilers helps considerably, but the inlet efficiency is still difficult to quantify. An isokinetic, isoaxial inlet would minimize inlet artifacts.

For small particles, inlet inefficiencies in the Berner and MOUDI have little effect. Mass median diameters determined by these two devices show good agreement despite the expected humidity-caused particle size changes expected within the impactors. However, the accumulation mode mass differed significantly during the Bellows intercomparison. The cause of this has not been determined. There is a possibility that SO_4^{-2} was not efficiently extracted from the aluminum impaction substrates.

The Sierra impactor appears to characterize the accumulation mode poorly, collecting less mass and indicating excessively large median particle diameters. If it is to be used to determine size distributions extending below 1 μm, an update of the Sierra calibration (Willeke, 1975) is needed to provide data on submicrometer particles and the effects of using Whatman 41 impaction substrates. It may be that its few, strongly cross-sensitive stages cannot yield enough information to give anything beyond a rough idea of ambient size distributions, particularly for the accumulation mode.

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