PIXE ANALYSIS OF MOUDI FILTERS

Eduard Stelcer, Anthony L. Morrison, Armand J. Atanacio and David D. Cohen
ANSTO Institute for Environmental Research, Sydney, Australia
Macquarie University Graduate School of the Environment, Sydney, Australia

Abstract

This paper will present results from a series of analyses performed on 8-stage Micro Orifice Uniform Deposit Im pactor (MOUDI) sampling substrates using accelerator-based Particle Induc ed X-ray Emission (PIXE) analyses. These experiments aimed to better understand the influence of aerosol deposition on each of the MOUDI stages on the PIXE analysis results.

Keywords: PIXE, MOUDI, Coverage Factor.

1. Introduction

Particle Induced X-ray Emission (PIXE) is a well established nuclear-based Ion Beam Analysis (IBA) technique used for multi-elemental analysis of aerosols collected on filters (Cohen, 1993, Cohen, 1998). This technique is based on counting the number of characteristic X-rays emitted from elements in the aerosols by bombardment with a high energy beam of charged particles from an accelerator.

When the aerosols are deposited on a filter as a thin uniform layer covering the entire collection area, *A* of the filter, the areal mass density, $m_a(Z)$ of an element, *Z* can be calculated from the measured X-ray yield. The total deposited mass, *m(Z)* of each element in collected aerosols can then be calculated from areal mass densities and the known deposition area, as follows:

$$m(Z) = m_a(Z) \times A \quad (1)$$

However, in the case of aerosols collected by MOUDI sampler, the aerosols are distributed on sampling substrates as small separated islands or spots Figure 1. Furthermore, the deposition pattern on the substrates is not uniform (Maenhaut *et al.*, 1993). As a result questions arise regarding the PIXE determination of areal mass densities and subsequent calculation of elemental mass concentrations.

MOUDI samplers are cascade impactors designed to sample size-selective aerosol particles. A MOUDI model 100 with 8 impactor stages was used in this study. It can accommodate 37mm or 47mm sampling substrates and according to the manufacturer, the particle deposit under the nozzles can be spread out over a 25mm diameter impaction area (MSP Corporation, 2007). According to Marple, (Marple *et al.*, 1991) it requires a 28mm diameter for the inlet and stage 1, and 27mm for all other stages. In the paper by (Maenhaut *et al.*, 1993) the estimated impaction area differed from stage to stage, ranging from 22.8mm in diameter for stage 2 to a maximum of 28.8mm in diameter for stage 7.

Visual inspection of subs trates Figure 1 confirmed the particle deposits do indeed spread out inside the 28mm diameter area for stages 1 to 8. This suggests that any strai ghtforward PIXE analysis should be perform by bombardment with a 28mm diameter beam or smaller if it can be shown to be representative of the aerosol deposits on substrates. For the inlet and exit stages, due to deposition covering most of the substrate area, the beam size should not be problematic as long as the aerosol deposition is uniform.

As bombardment with the 28mm diameter beam is often impractical in most PIXE laboratories, we investigated the possibility of straightforward PIXE analysis with smaller diameter beams.

2. Method

For sampling substrates, we used stretched Teflon filters of 47mm diameter. One set of filters was Apiezon grease-coated and artificially loaded with aluminium silicate. The second set consisted of uncoated filters and had been used for collecting ambient aerosols at a Hunter Valley site (100km North-East from Sydney). Each filter was then analysed for elemental concentrations by PIXE using 8, 12, 16, 20, 22, 24, 26 and 28mm diameter proton beams of 2.6MeV energy.

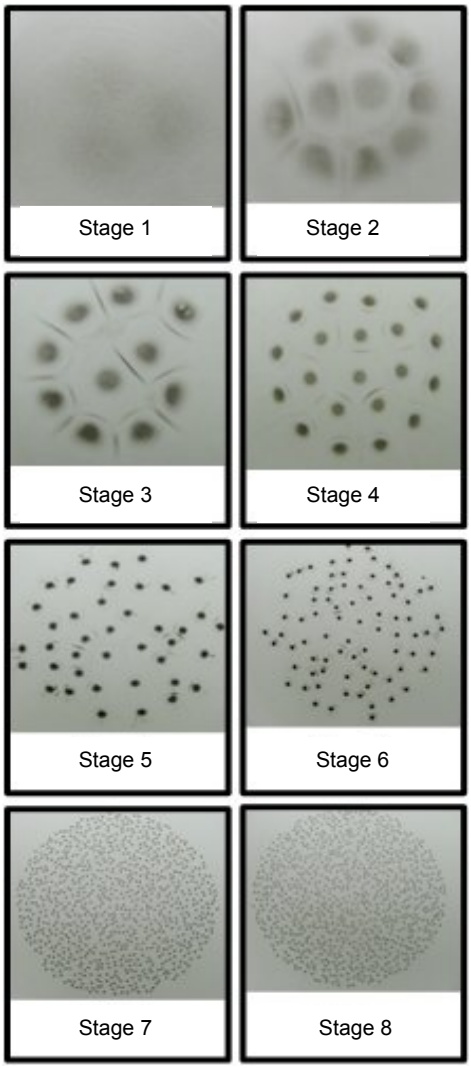

Figure 1. Deposition patterns of aerosols on MOUDI sampling substrates.

3. Results and discussion

3.1. Areal mass densities of the elements

As expected, PIXE analyses performed with different beam sizes produced different results. It can be seen from Figure 2 that the differences in analysed areal mass densities were much higher for analysis with 8, 12, 16 and 20mm diameter beams than for analysis with 22, 24, 26 and 28mm diameter beams.

The standard deviations for average areal mass densities of the dominant elements analysed using 22, 24, 26 and 28mm diameter beams are presented in Table 1. It can be seen that the standard deviations resulting from the deposition of aluminium silicate on coated substrates were within 5% of the analysed average values. These standard deviations were therefore within 5% error values

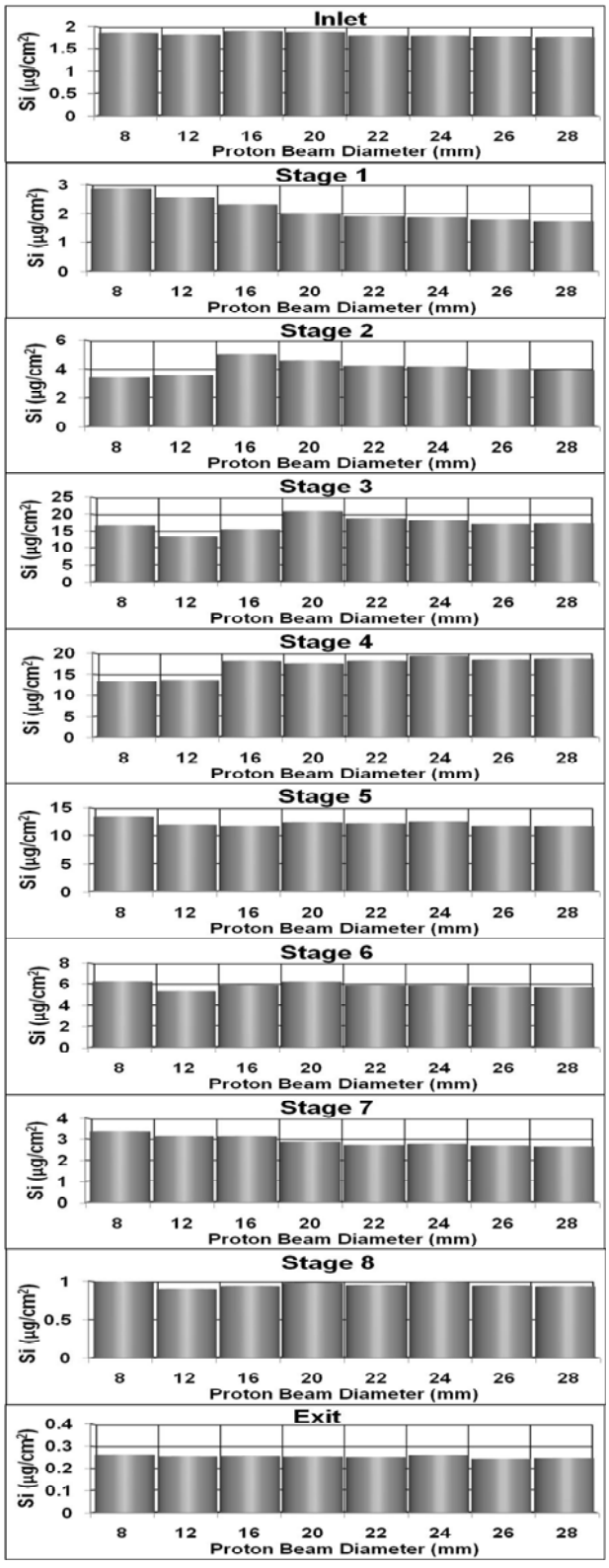

Figure 2. Areal mass densities for Si in aluminium silicate for each MOUDI stage. Analysed with proton beams of 8, 12, 16, 20, 22, 24, 26 and 28mm in diameter.

Table 1. Standard deviations for average areal mass density for dominant elements in aluminium silicate and ambient aerosols. Analysed with 22, 24, 26 and 28mm beams.

Proton beams: 22, 24. 26 and 28mm			Stage 1	Stage 2	Stage 3	Stage 4	Stage 5	Stage 6	Stage 7	Stage 8
Standard deviation (%)	Aluminium Silicate (coated filters)	Al	3.2	4.6	4.6	2.9	3.4	1.5	4.0	5.0
		Si	4.4	3.2	4.4	2.5	3.2	1.6	1.9	2.5
Standard deviation (%)	Ambient Aerosols (not- coated filters)	Al	7.2	8.7	7.1	3.6	9.2	5.4	22.6	27.0
		Si	6.2	7.8	4.0	6.3	3.5	2.4	3.4	1.5
		S	3.2	6.0	4.0	4.1	3.1	1.4	2.5	2.0
		K	6.4	8.6	4.5	5.9	5.7	9.9	20.4	19.8
		Fe	7.6	6.1	4.3	7.9	8.6	11.9	13.8	21.6

for certified Micrometer foil standards used in the PIXE system calibration. This indicates that straightforward PIXE analysis of grease coated MOUDI substrates is not limited to bombardment by a 28mm diameter beam but can also be performed with any beam where the diameter is not smaller than 22mm.

For PIXE analysis using 8, 12, 16 and 20mm diameter beams, areal mass density correction factors, *F* are needed for MOUDI stages 1 to 8. For the Inlet and exit stages, correction factors are not required as the impacting aerosols are deposited reasonably uniformly over the entire substrate surface. The areal mass density correction factors estimated empirically are listed in Table 2.

Correlations between corrected areal mass densities for MOUDI stages 1 to 8 from analysis with a 16mm diameter beam (for Si and Al in aluminium silicate) and average areal mass concentrations calculated from analyses with 22, 24, 26 and 28 mm diameter beams are presented in Figure 3.

Table 2. Areal mass density correction factors, *F* for analysis with 8, 12, 16 and 20mm proton beams.

Proton beam diameter (mm)	Correction factor, *F*
8	1.095 ± 0.002
12	1.229 ± 0.008
16	1.049 ± 0.004
20	0.943 ± 0.002

With respect to uncoated substrates used for collecting ambient aerosols, standard deviations for analysed areal mass densities in some cases were much higher than 5%. This was probably due to a bounce-off effect during the impaction process and indicates that the coated substrates are preferable for MOUDI samplers compared to uncoated substrates.

3.2. Mass concentrations of the elements

In a typical aerosol analysis, the calculation of total deposited mass for elements analysed by PIXE requires determination of the elemental areal mass densities and the total area of the aerosols deposited on the sampling substrate.

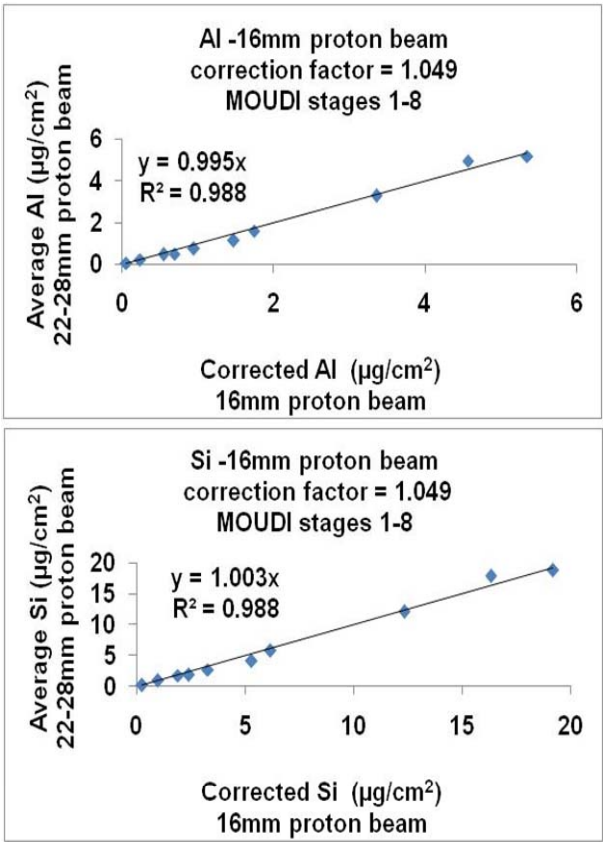

Figure 3. Correlations between corrected and average areal mass concentrations for Si and Al in aluminium silicate for MOUDI stages 1 to 8 analysed with 16mm proton beam.

However, with regards to MOUDI sampling substrates, aerosols tend to be distributed as separated islands or spots as shown in Figure 1. Examination under the optical microscope Figure 4 shows that due to the nozzle spraying effect, the area of deposited spots on the substrates cannot be accurately determined. Collisions between particles following the air stream out of the impaction region from adjacent nozzles results in additional particle deposition in the spaces between spots Figure 1. All these factors make estimating the total deposition areas extremely difficult.

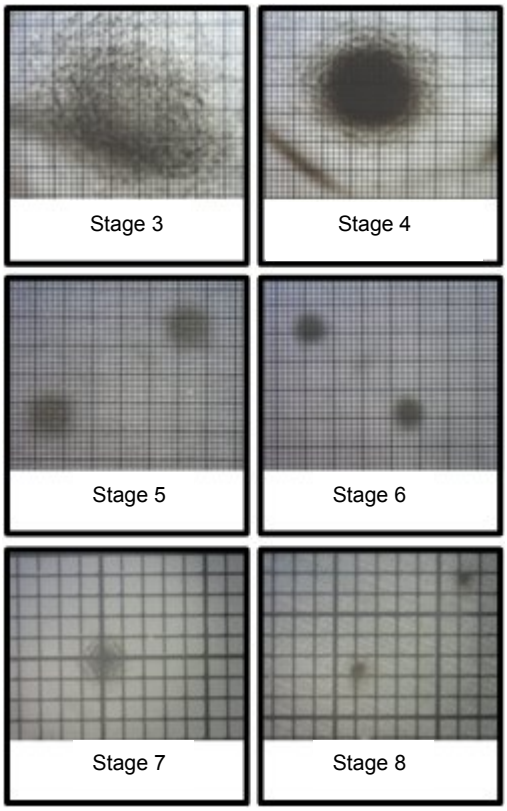

Figure 4. Image of deposited spots on MOUDI sampling substrates under optical microscope. Distance between grids is 0.1 mm.

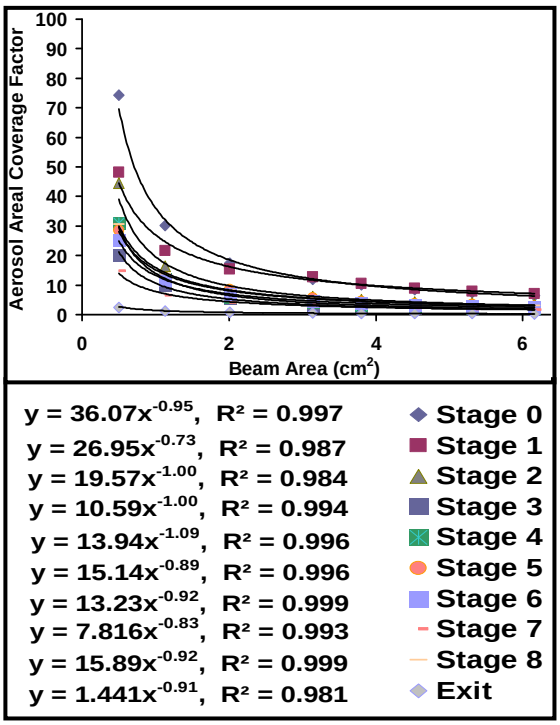

Figure 5. Relations between beam areas and aerosol areal coverage factors for each MOUDI stage.

Furthermore, only some of the protons from incoming beam will interact with the deposited aerosols and the measured areal mass determined from PIXE will be lower than its true value.

To bypass these problems and obtain a more accurate estimation of the total deposited mass of elements on the substrate, we utilised the aluminium silicate gravim etric mass, the atomic mass of elements in aluminium silicate, exposed filter area to proton beam and the measured areal densities of those element s by PIXE to calculate the aerosol areal coverage factor, *C* for each beam diameter and for each MOUDI stage.

X-Ray Diffraction analysis (XRD) showed that the aluminium silicate was composed from Zeolite ($Al_{2.8}Si_9O_{22}$) and traces of Heulandite ($K_{1.5}Ca_{4.5}Al_{8.5}Si_{28}O_{72}$). Due to the negligible amount of Heulandite in the mixture only Zeolite was used for the elemental concentration calculations. The calculated aerosol areal coverage factors for each beam diameter and for each MOUDI stage are listed in Table 4. Relations between beam areas and calculated aerosol areal coverage factors are presented in Figure 5.

The total deposited mass, $m(Z)$ of an element, Z in collected aerosols can be now estimated from PIXE measured aerial density, $m_a(Z)$ as follows:

$$m(Z) = m_a(Z) \times S \times F \times C \quad (2)$$

where S is the applied beam area, F is the areal mass density correction factors from Table 2, and C aerosol areal coverage factor from Table 4.

Areal mass density correction factors are not required for any of the stages when the 22, 24, 26 and 28mm diameter proton beams are used; neither are they required for analysing the inlet and exit stages when smaller sized beams can be used. However, it should be noted that the aerosol coverage factor values, C for the exit stage are only estimates as the sample weight was insuf ficient for more accurate determination at this time.

Figure 6 shows the correlation between estimated total deposited mass and theoretically calculated mass for aluminium and silicon in the deposited aluminium silicate for 8mm diameter proton beam and Figure 7 for 22mm diameter proton beam.

Table 4. Aerosol areal coverage factors, *C*.

Stage	8mm	12mm	16mm	20mm	22mm	24mm	26mm	28mm
Inlet	74.4 ± 3.0	30.2 ± 2.3	17.4 [a]	12.0 ± 0.7	10.1 ± 0.3	8.5 ± 0.3	7.5 ± 0.4	6.4 ± 0.3
1	48.2 ± 1.8	21.7 ± 1.0	15.4 ± 0.4	12..8 ± 0.6	10.4 ± 0.4	9.0 ± 0.4	7.9 ± 0.3	7.0 ± 0.2
2	44.6 ± 2.4	16.5 ± 0.4	7.8 ± 0.3	6.1 ± 0.3	5.1 ± 0.2	4.4 ± 0.1	3.9 ± 0.2	3.4 ± 0.2
3	20.2 ± 0.7	10.1 ± 0.4	5.7 ± 0.2	3.0 ± 0.1	2.6 ± 0.1	2.3 ± 0.1	2.1 ± 0.1	1.7 ± 0.1
4	30.9 ± 0.9	11.9 ±0.4	5.9 ± 0.2	4.3 ± 0.2	3.3 ± 01	2.6 ± 0.1	2.3 ± 0.1	2.0 ± 0.1
5	28.6 ±1.2	12.6 ±0.5	8.5 ± 0.4	5.7 ± 0.2	4.5 ± 0.2	3.7 ± 0.2	3.4 ± 0.2	2.9 ± 0.1
6	25.3 ± 0.9	11.5 ± 0.2	6.9 ± 02	4.7 ± 0.2	3.9 ± 0.2	3.3 ± 0.1	2.8 ± 0.1	2.5 ± 0.1
7	14.8 ± 0.5	6.4 ± 0.3	4.1 ± 0.1	3.2 ± 0.1	2.7 ± 0.1	2.2 [a]	1.9 [a]	1.7 ± 0.1
8	30.6 ± 0.4	13.7 ± 1.8	8.4 ± 0.9	5.7 ± 0.6	4.5 ± 0.4	3.9 ± 0.6	3.4 ± 0.4	3.0 ± 0.4
Exit	7.5 [b]	3.8 [b]	2.6 [b]	1.8 [b]	1.1 [b]	1.1 [b]	1.1 [b]	1.1 [b]

[a] Errors are less then 0.1. [b] Estimated at this time as sample weight was insufficient for more accurate determination.

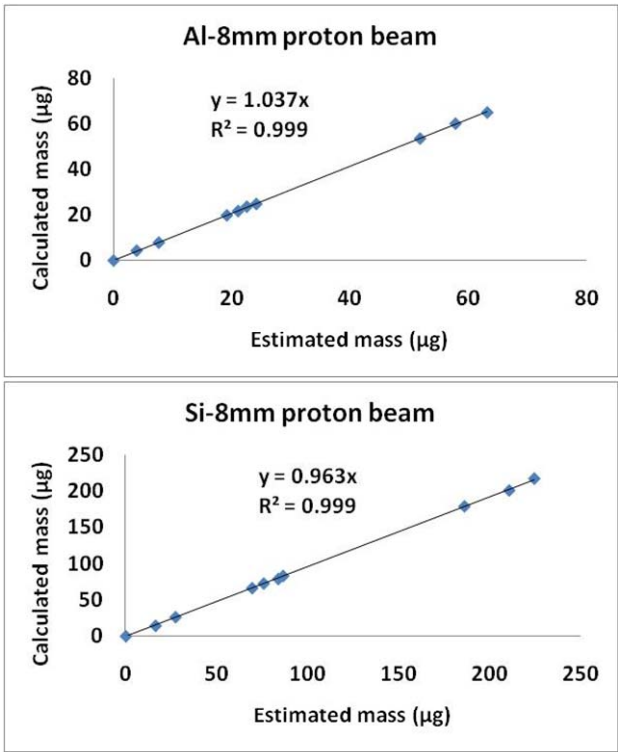

Figure 6. Correlation between estimated total deposited mass and theoretically calculated mass for aluminium and silicon for 8mm proton beam.

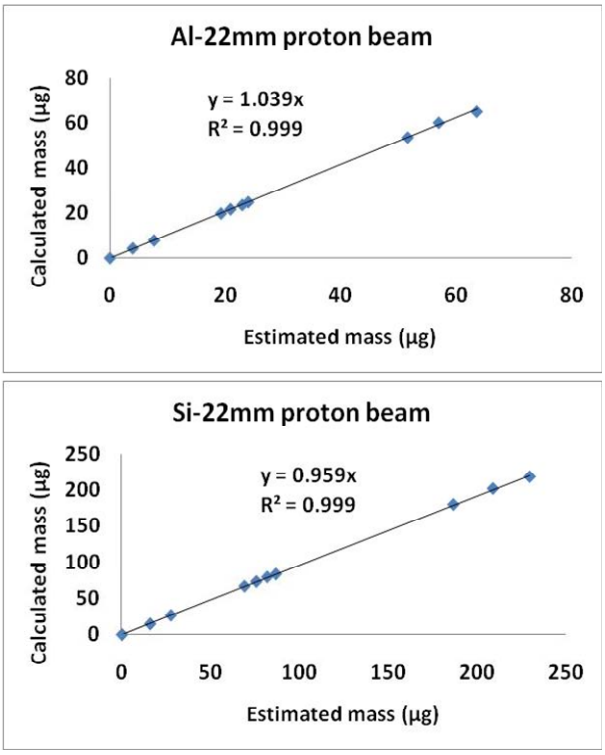

Figure 7. Correlation between estimated total deposited mass and theoretically calculated mass for aluminium and silicon for 22mm proton beam.

4. Conclusions

PIXE is an established powerful nuclear tool for aerosol filter analysis. However, airborne particulates collected using MOUDI samplers are deposited randomly as non-uniform spots on each substrate stage. As a result, determining both areal mass densities and the related calculation of elemental mass concentrations using standard PIXE aerosol analysis methods may be unreliable. We have presented a modified PIXE analysis method incorporating the use of areal mass density correction f actors, *F* and aerosol areal coverage factors, *C* to successfully analyse aerosol substrates with non-uniform deposits that result from the use of MOUDI samplers.

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MSP Corporation *Micro-Orifice Uniform Deposit Impactor (MOUDI™) and Nano-MOUDI™*. Available online at: http://www.mspcorp.com.