

A low-cost particle counter as a realtime fine-particle mass monitor

Cite this: *Environ. Sci.: Processes Impacts*, 2013, **15**, 433

Amanda L. Northcross,^{*a} Rufus J. Edwards,^b Michael A. Johnson,^c Zhong-Min Wang,^d Kunning Zhu,^d Tracy Allen^e and Kirk R. Smith^a

Exposure to particles with aerodynamic diameters less than 2.5 μm is estimated to cause significant morbidity and mortality worldwide leading many countries to develop ambient air pollution standards and guidelines. At local scales, community and environmental justice groups are also concerned about $\text{PM}_{2.5}$ concentrations that may be elevated above regional concentrations typically measured by centrally located monitors and standards as well. In an attempt to develop a low cost, easy to use monitor we evaluated a low-cost optical particle counter, the DylosTM, as a fine particulate mass sensor. Modified into a system called the Berkeley Aerosol Information Recording System (BAIRS), we compared performance against standard commercial instruments in chambers using polystyrene latex spheres, ammonium sulphate, and woodsmoke and in an urban ambient setting. Overall we find that the limit of detection of the BAIRS is less than 1 $\mu\text{g m}^{-3}$ and the resolution is better than 1 $\mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$. The BAIRS sizes small (<0.5 μm) particles, and is able to accurately estimate the mass concentration of particles of varying composition including organic, inorganic, and ambient particles. It is able to measure concentrations up to 10.0 mg m^{-3} . In an ambient roof-top test of the BAIRS and a more expensive commercially available light scattering particle monitor the BAIRS response tracked well with the commercial monitor and daily means were within 80% of each other. We conclude that with appropriate modification the system could be developed into an accurate low cost realtime particle mass monitor for use in a wide range of applications.

Received 13th July 2012
Accepted 6th November 2012

DOI: 10.1039/c2em30568b

rsc.li/process-impacts

Environmental impact

This study aims to evaluate a low cost particle monitor for use as an ambient particle monitor. As monitors that are currently used are too costly to be used to support large-scale epidemiological studies as well as community groups wanting to better characterize exposures locally, we evaluated a low-cost commercial device designed for monitoring. Development of this monitor will be able to meet the needs of both groups.

Introduction

Combustion in its many forms is a large source of health-damaging air pollution in the US and globally. According to the WHO Comparative Risk Assessment, urban ambient air pollution from fine particles is responsible for 0.8 million premature deaths each year annually world-wide¹ and particles from the combustion of household solid fuels such as biomass and coal are estimated to be responsible for more than 1.6 million premature additional deaths annually around the world.² Most health effect studies have relied on 24 h or annual mean mass

concentrations,^{3,4} although there are efforts to understand effects using other metrics for example short-term peak levels or particle number. The most commonly used measure of major health impacts from combustion-derived air pollution is the particle mass concentration.⁵ Health-based standards throughout the world are increasingly specifying limits based on particles less than 2.5 μm ($\text{PM}_{2.5}$), but some studies seem to indicate potential benefits of measuring even smaller sizes, perhaps particles less than 1.0 μm ($\text{PM}_{1.0}$).^{6,7} Combustion particles are nearly all in the size range of 2.5 μm or smaller.⁸

Epidemiological studies using PM measurements at ambient centrally located monitoring stations routinely show consistent effects for important health endpoints.^{4,9,10} However human exposures may differ substantially in temporal and spatial distribution from what these stations show. Evidence from Los Angeles based on improved exposure assessment modeling methods utilizing land use regression (LUR) indicated that chronic exposure to particles in urban environments may result

^aDepartment of Environmental Health Sciences, School of Public Health, University of California Berkeley, Berkeley, CA, USA. E-mail: amandaln@gmail.com

^bEpidemiology, School of Medicine, University of California Irvine, Irvine, CA, USA

^cBerkeley Air Monitoring Group, Berkeley, CA, USA

^dEnvironmental Health Laboratory, California Department of Public Health, Richmond, CA 94804, USA

^eElectronically Monitored Ecosystems, 2229 Fifth Street, Berkeley, CA 94710, USA

in health effects two to three times greater than earlier believed.^{11,12} Evaluation of LUR models developed using stationary PM monitors shows that the majority of the variance of estimated personal exposures for PM is explained by temporal changes emphasizing the need for time resolved PM data.¹³ Increasing concern exists for populations with systematically elevated exposures as a result of local point sources in urban environments, which is difficult to capture without exposure assessment which includes both temporal and spatial resolution at scales much smaller than current central site monitoring allows. To better understand the relationship of particle exposures and ill-health and to better target control measures to address exposure, there is a need for technologies to monitor particle levels in a wide range of locations, preferably with high temporal resolution.

Although scientific research is an important driver of air pollution policy, community groups and other members of the public have also been important in bringing this issue to the forefront and ensuring that the policy protects communities with limited voices, but who are commonly the most impacted.¹⁴ The current costs and/or complexity of commercial instruments limits the ability such groups have for monitoring pollution in their communities. However these groups commonly do not have access to the instrumentation to measure the distribution of PM. To address this need for instrumentation to support large-scale epidemiological studies as well as community groups wanting to better characterize exposures locally, we evaluated a low-cost commercial device designed for monitoring.

The goal of this evaluation is to determine the accuracy, sensitivity, measurable range, linear response and resolution of a less-expensive monitoring system taking advantage of a low-cost particle-counting sensor in an existing commercial device. This work follows from previous success in adapting commercial smoke detector technology for monitoring in high particle environments in developing countries.¹⁵ Here we report investigations of how well the modified device works in comparison to standard monitoring approaches.

Technical design

The Dylos Air Quality Monitor™ is designed, manufactured, and distributed by the Dylos Corporation (Riverside California). Called here simply the Dylos, it was designed for use in homes and offices to monitor particle number concentrations. Stated examples (<http://www.dylosproducts.com>) include testing the efficiency of in-home air cleaners, or monitoring particulate concentrations in woodworking shops. Our interest in the Dylos is due to its low cost, <\$300, in comparison to the existing real-time particle mass monitors (>\$2000).

The Dylos uses a small computer fan to draw in air and particles and funnels them through baffles molded into the case past the laser beam operating at 650 nm wavelength. The wide air path allows the low pressure fan to draw a relatively large flow rate, which does not need to be calibrated. A photo-diode is located close to the scattering volume, positioned so that it captures scattered light from many angles. There are no lenses,

mirrors or other focusing optics commonly used in other optical particle monitors, which reduces cost and the interference that can occur from particles coating the optical surfaces.

The model we used reports particle numbers in two size bins: PM_{0.5} which measures particles sized 0.5 μm and greater, and PM_{2.5} which measures particles sized 2.5 μm and greater. The instrument does not use a physical size selector such as an impactor or cyclone, but conducts the size discrimination using an algorithm on the signal from scattered light. The flow rate is not monitored, it is inferred from a patent of the manufacturer that the fluctuations are compensated using the measured width of the peaks from the light scattered from the particles.¹⁶ The focus of our work is to evaluate the ability of a modified version of the Dylos, as an ambient particle mass monitor, relying only on minimal data processing. For simplicity, we call this prototype the BAIRS (Berkeley Aerosol Information System).

Changes made

The Dylos sold by the manufacturer did not meet all the requirements needed for an ambient real-time particle mass monitor, we made several modifications.

Particle bin size

In order to investigate the ability of the Dylos to distinguish between particles of differing sizes we requested the manufacturer to add two additional size bins. These bins were uncalibrated and reported particles sizes between 0.5 and 2.5 μm. The two intermediate bins are based on a measure of the peak height in the raw voltage from the photodetector, not particle size directly, but are expected to correlate with size.

Data logging

We also required larger storage and a precise time resolution than provided by the commercial device. The company provided us with modifications of their standard firmware protocol, which allowed requests for data from all four channels at consistent time intervals and thus the use of our own datalogger.

The Owl™ datalogger (<http://www.emesystems.com/OWL2pepr.htm>) has been incorporated into the BAIRS through external connection to the 9 pin COM Port. It records the processed signal from the photodiode as particles per standard cubic foot (scf), as sent by the Dylos. The datalogger can be programmed to record data at any time interval. It also logs temperature data as well as data from other sensors and allows for a variety of communication options. The BAIRS responds to requests for data at an interval set by the datalogger, which allows better time resolution in comparison to the standard Dylos.

Battery life

The standard Dylos is powered by line current and demands too much power for the convenient use of batteries. At our request, the manufacturer provided a more efficient, quieter fan and commands that permit external control of the display screen back light and the power for the entire instrument. This allows for lower overall power consumption as well as non-continuous

sampling schemes. The BAIRS also incorporates modifications designed to accommodate a rechargeable battery.

Experimental methods

All experiments conducted in this study are summarized in Table 1.

Chamber samples

An indoor aerosol chamber was utilized to sample various aerosols in a controlled and repeatable setting. The $\sim 1 \text{ m}^3$ chamber is lined in aluminum and has an adjustable side to allow for filter sampling while maintaining the atmospheric pressure. Aluminum was used for ease of cleaning, to reduce chemical wall reactions and to minimize electrostatic effects. The chamber is equipped with a TSI DustTrak™ (model 8520, Shoreview, MN) for continuous particle monitoring as well as model PCXR8 SKC (Eighty Four, PA) personal sampling pumps and BGI Triplex Cyclone™ (Waltham, MA) to achieve a cut point of either 2.5 or 1.0 μm for gravimetric filter sampling. Flow rates were calibrated for either 1.5 or 3.5 lpm respectively, immediately before and after sampling using a Gilian Gilibrator™ (Sensidyne Mülheim, Germany). At the start of each experiment, a low particle environment was created in the chamber using an air compressor connected to a HEPA filter.

Particles were added to the chamber using two methods. Ammonium sulfate (0.01 M $(\text{NH}_4)_2\text{SO}_4$) and polystyrene latex spheres were introduced into the chamber using a medical grade nebulizer, mean particle diameter (0.5–0.7 μm), in series with a diffusion dryer. Combustion particles were also tested by burning a predetermined mass of wood in the chamber burn pot. Particle concentration was controlled by adding clean air while concurrently removing the same volume of air using a sampling pump while smoke was added. A small mixing fan ran during each experiment to ensure complete mixing in the chamber. The BAIRS was placed inside the chamber. The DustTrak and pump–filter combination were placed outside but connected *via* a sampling tube.

Ambient samples

Ambient particulates were sampled to evaluate the ability of the BAIRS to perform within an outdoor setting with real atmospheric aerosols. All ambient samples were conducted on the roof of the State of California Health and Human Services Air Pollution Lab in Richmond, CA with a co-located DustTrak and an E-bam beta attenuation monitor (Met-One™). The DustTrak

had a $\text{PM}_{2.5}$ impactor as a particle size selector, and the E-bam was affixed with a cyclone to measure $\text{PM}_{2.5}$.

Results and discussion

Laboratory tests

Polystyrene latex spheres. The BAIRS's ability to distinguish fine particles was tested using polystyrene latex (PSL) spheres in the indoor smoke chamber. The PSL spheres were 0.490 μm calibration standard spheres. A mono-dispersed particle distribution is used to display the ability of the device to correctly size particles at concentrations that are relevant for the proposed application. In Fig. 1, the 0.5 μm sized bin shows the largest response to the PSL spheres as is expected. At 1×10^7 particles per m^3 the BAIRS isolates the PSL spheres in the 0.5 μm bin, and the other sizes do not respond to the PSL spheres. At higher concentrations the larger size particle bins also showed a response to the PSL spheres, the largest of which is 2 orders of magnitude less and negligible in comparison to the response of the 0.5 μm sized bin. The maximum number concentration of the PSL spheres in the chambers was 3.2×10^7 particles per m^3 , much higher than expected in an ambient setting. For example researchers at Clarkson University¹⁷ monitored particle number concentrations in New York and reported a mean concentration of $\sim 1 \times 10^6$ particles per cm^3 in the 0.1–0.47 μm particle size range. Although some agglomeration/accumulation of PSL spheres would be expected, response from the three larger size bins is most likely caused by optical particle counters counting the scattering by more than one particle in the laser path as one larger particle as opposed to several smaller particles.¹⁸

Sensitivity to different aerosols. The BAIRS, like all light-scattering instruments, can respond differently to particles of varying compositions. In comparison to the DustTrak the relative response between the two instruments changes as the particle source changes (Fig. 2). The differing slopes suggest that either the DustTrak or the BAIRS has a varying sensitivity to the composition or particle size. The PSL spheres are the smallest aerosols at 0.49 μm tested, the approximate mean particle diameter of the woodsmoke and the ammonium sulfate are $\sim 0.7 \mu\text{m}$ and $0.8 \mu\text{m}$ respectively.⁸ The refractive index for PSL spheres is 1.58, woodsmoke is 1.57, and ammonium sulphate is 1.53.¹⁹ The ability of particles to scatter light is dependent on both particle size and composition and the differing sensitivity is caused by both factors. In this study it appears that the difference in sensitivity for these aerosols is more closely related to the refractive index than the particle size. The refractive indices for woodsmoke and PSL spheres are much closer as are the slopes, while that of ammonium sulphate is lower and the slope is higher. These results are not definitive as this is a comparison between instruments.

Mass concentration.

$$m(d_p) = \frac{4}{3} \pi d_p^3 \rho_p n(d_p) \quad (1)$$

To convert the number concentration $n(d_p)$ into a mass concentration $m(d_p)$ as a function of particle diameter, eqn (1) is

Table 1 Summary of experimental runs conducted

Aerosol type	Location	Mass concentration range	Number of runs
Woodsmoke	Chamber	0–1200 $\mu\text{g m}^{-3}$	4
0.5 μm polystyrene latex spheres	Chamber	0–190 $\mu\text{g m}^{-3}$	2
Ammonium sulphate (crystalline)	Chamber	0–170 $\mu\text{g m}^{-3}$	4
Ambient aerosol	Roof top	0.8–39 $\mu\text{g m}^{-3}$	4

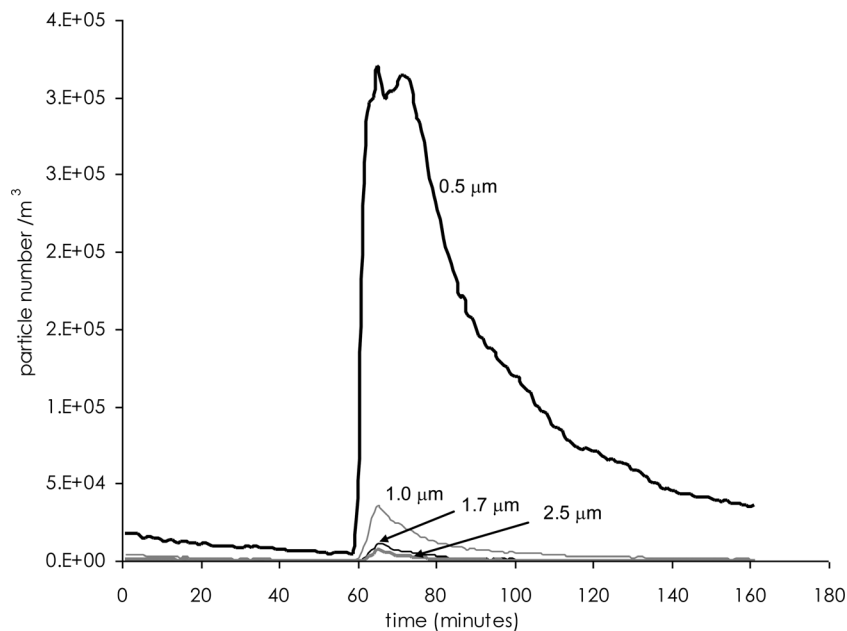


Fig. 1 Particle number concentration of PSL spheres in the chamber. The 0.5 μm and 2.5 μm channels are factory calibrated, while 1.0 μm and 1.7 μm are estimated.

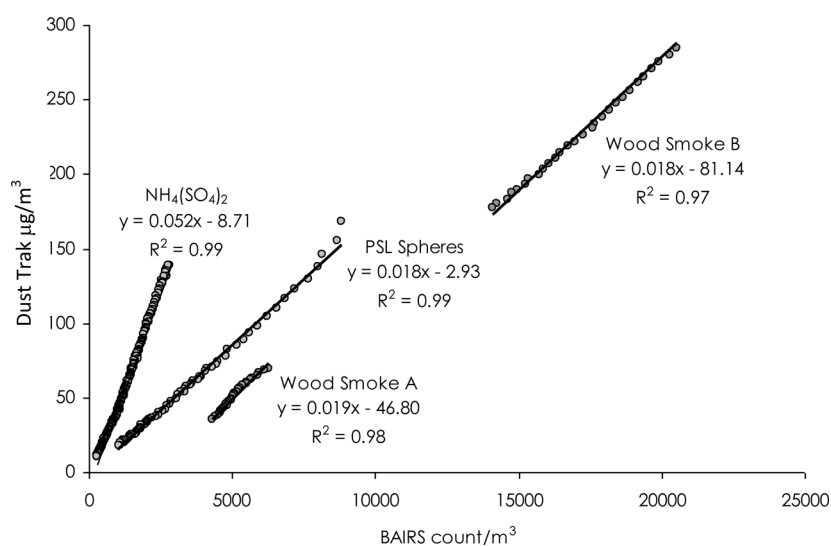


Fig. 2 BAIRS versus DustTrak™ chamber experiments for ammonium sulfate and woodsmoke for varying concentrations. The BAIRS number is the count of particles in the range of 0.5 μm to 2.5 μm ; computed from the raw data by subtracting the >2.5 count from the >0.5 count.

used, where ρ_p is the particle density and d_p is the mean particle diameter. In the indoor chamber, a single source of particles was used, and the physical and chemical compositions of these aerosols were known. The nebulizer used to generate the inorganic aerosols produced particles with a mean aerosol diameter of ~ 0.7 μm , and the PSL spheres were of a single diameter. The particle density of the inorganic aerosols was estimated using the AIM online model,¹⁹ the model requires solution molar concentration and ambient temperature and humidity. Woodsmoke has been well characterized for both composition and particle size, however in the case of ambient aerosols much less was known. In the absence of a known particle size distribution,

the aerosol mass was calculated for each particle size bin. The BAIRS measures the particle number concentration at four different particle sizes (0.5, 1.0, 1.7, 2.5 μm). Only two of the size bins (0.5 and 2.5 μm) are calibrated by the manufacturer. In order to estimate the mass concentration the mean particle size of each bin (0.75, 1.4, 2.1 μm) was used to estimate the mass concentration (density = 1.0 g cm^{-3}).

The DustTrak measured aerosol concentration was adjusted with the mass concentration from gravimetric filter sampling for the chamber experiments and a federal approved reference standard Met-One E-bam for the ambient samples. The ratio of the concentration determined by gravimetric concentration or

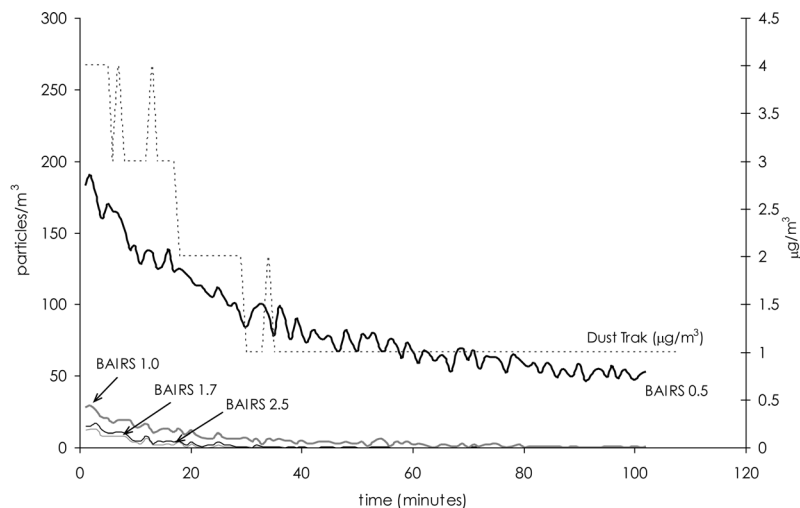


Fig. 3 BAIRS and DustTrak™ in a low particle environment. BAIRS number counts reported for calibrated sizes of 0.5 and 2.5 μm and larger and uncalibrated sizes of ~ 1.0 and 1.7 μm and higher. The DustTrak™ mass concentration was not adjusted by gravimetric filter measurement.

the E-Bam to the mean concentration measured by the Dust-Trak for the same time period was used to adjust the DustTrak minute scale data.

Limit of detection. The limit of detection (LOD) is defined as 3 times the standard deviation of the instrument signal during sampling in a near-zero particle environment. The chamber was filled with filter room air to create near-particle-free air. The limit of detection of the BAIRS is 16 particles per m^3 or a mass concentration of $0.1 \mu\text{g m}^{-3}$ ($5.02 \times 10^{-4} \mu\text{g}$ per particle) assuming a mean particle size of 1.0 μm and a density of 1.2g cm^{-3} . Fig. 3 shows the number concentration for the BAIRS, and the un-adjusted mass concentration for the DustTrak. The resolution of the BAIRS is much finer than the DustTrak, and it has a lower limit of detection. The Dylos does not measure less than 1 particle; however the unit conversion from scf to m^3 creates numbers less than 1.

Concentration dependence. Fig. 2 shows the particle number concentration from the BAIRS compared to the adjusted mass concentration from the DustTrak using varied concentrations of woodsmoke in the indoor chamber. Fig. 2 shows the linear relationship at both high and lower concentrations of woodsmoke. At both levels of smoke concentrations the slope is almost the same suggesting that the response between the BAIRS and DustTrak does not change between the two different concentrations of the same type of aerosols.

Ambient testing

The E-bam was used to adjust the readings from both the DustTrak and the BAIRS to provide a direct comparison between the ability of the two instruments to measure $\text{PM}_{2.5}$ in an outdoor environment. The correction factors for both the DustTrak and the BAIRS using the E-Bam were determined using data for the entire sampling period. The composition of ambient aerosols can change temporally due to varying sources,

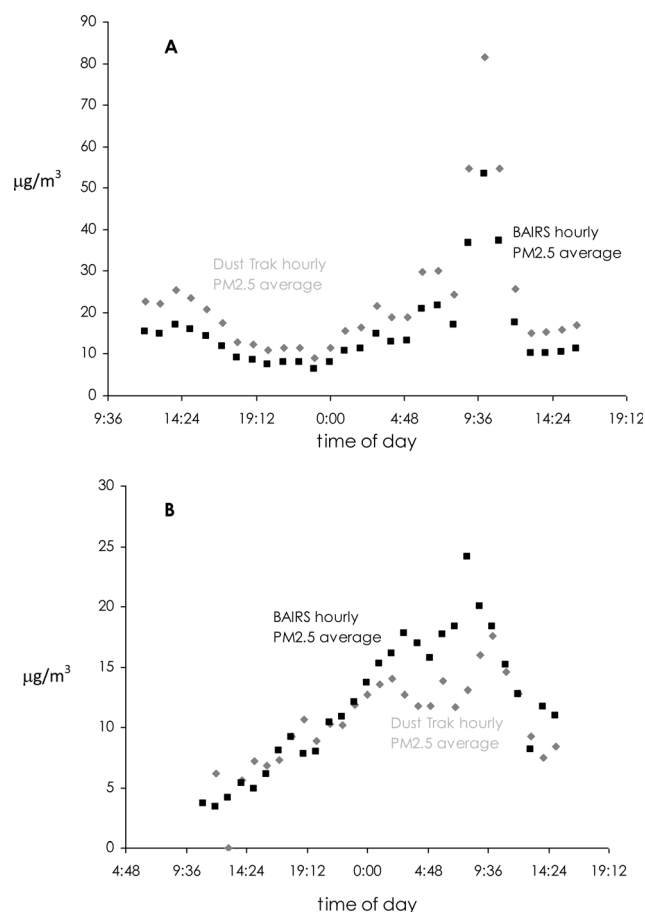


Fig. 4 E-bam calibrated minute by minute results for two sampling periods. (A) Period 1. DustTrak™ adjusted, BAIRS adjusted. (B) Period 2. DustTrak™ adjusted, BAIRS-adjusted, mean particle diameter.

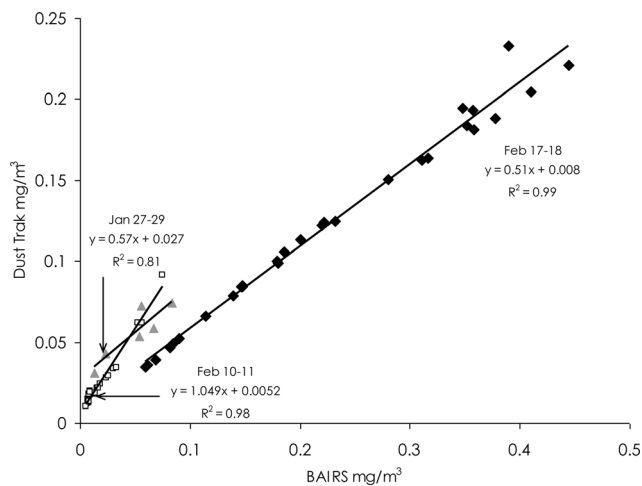


Fig. 5 Linear regression between DustTrak™ and BAIRS for ambient particle sampling for three periods, each period for at least 24 hours. Both BAIRS and DustTrak™ measurements adjusted by the E-Bam 24 hour measurement.

which can affect the particle optical properties and the response of the instruments. The mean correction factor over the entire sampling period allows the temporal changes in the particle optical properties to be averaged out over the entire sampling period. This is the correction method commonly used to correct optical aerosol monitoring instruments for various types of aerosols. Testing was conducted for a total of 110 hours over three weeks.

Fig. 4 shows the hourly averaged data from the DustTrak and the BAIRS collocated for two different ambient monitoring periods. The two instruments track closely; reporting almost the same values over both sampling periods. Period 1 had clean air as it had rained immediately prior to the sampling period and the humidity remained very high during the entire sampling period. Period 2 had higher concentrations and the reported values were close. The average concentration for period 1 was $11.9 \pm 0.3 \mu\text{g m}^{-3}$ and $11.1 \pm 0.2 \mu\text{g m}^{-3}$ using $\alpha = 0.05$ to calculate the confidence intervals. Period 2 had a mean average concentration of $25 \pm 8 \mu\text{g m}^{-3}$ and $32 \pm 6 \mu\text{g m}^{-3}$ for the BAIRS and DustTrak respectively.

Fig. 5 shows the hourly mean mass concentrations from the two instruments plotted against each other. The r^2 values are lower in comparison to Fig. 2 where the particle number was plotted versus the DustTrak mass for woodsmoke in the chamber on a minute-by-minute basis. Ambient aerosols are of varying composition and also vary temporally. Optical instruments respond differently depending on the aerosol composition. In the chamber test a single aerosol was used reducing the variability in the response to the aerosols producing higher r^2 values.

Conclusion

The BAIRS monitor represents a promising step towards creating a low-cost realtime particle mass monitor for use in field conditions. It is able to accurately “size” particles and

operate at a wide range of particle concentration. The BAIRS tracks well with a commercially available monitor and provides similar results. The resolution of the BAIRS is finer than the commercially available monitor, see Fig. 3, and the limit of detection is less than $1 \mu\text{g m}^{-3}$. Additional improvements are required to reduce power consumption making the device more portable with the addition of a battery pack and reducing the physical size by redesigning the case. In addition an operating software program will be needed to improve the efficiency of data processing. Conversion from measures of particle number to an estimate of mass requires assumptions about the characteristics of the particles being monitored. This is a drawback not unique to the BAIRS, however, but for all optical particle monitors. Nevertheless, our results indicate that the approach of adapting a low-cost particle counter into a system to measure the particle mass could potentially provide a portable device to accurately estimate the fine particle mass in a wide range of settings.

Acknowledgements

Sincere thanks are given to the California Department of Public Health, Environmental Health Laboratory and the California Air Resources Board (Agreement # 06-321) for financial and technical support. In addition we thank Marvin So for laboratory support and the STEER program (NIEHS: for his financial support). Although we are grateful for the technical input from Roger Unger the designer of the Dylos Particle Monitor, we received no financial or other support from the company.

References

- 1 A. J. Cohen, H. R. Anderson, B. Ostro, K. D. Pandey, M. Krzyzanowski, N. Künzli, K. Gutschmidt, C. A. Pope III, I. Romieu and J. M. Samet, *Others Urban air pollution. Comparative Quantification of Health Risks: Global and Regional Burden of Disease Attributable to Selected Major Risk Factors*, World Health Organization, Geneva, 2004, pp. 1353–1433.
- 2 K. R. Smith, S. Mehta and M. Maeusezahl-Feuz, *Indoor Air Pollution from Household Use of Solid fuels. Comparative Quantification of Health risks: Global and Regional Burden of Disease Attributable to Selected Major Risk Factors*, 2004, vol. 2, 1435–1493.
- 3 S. J. Dutton, B. Rajagopalan, S. Vedal and M. P. Hannigan, Temporal patterns in daily measurements of inorganic and organic speciated $\text{PM}_{2.5}$ in Denver, *Atmos. Environ.*, 2010, **44**, 987–998.
- 4 F. Laden, L. M. Neas, D. W. Dockery and J. Schwartz, Association of fine particulate matter from different sources with daily mortality in six US cities, *Environ. Health Perspect.*, 2000, **108**, 941–947.
- 5 WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide; Global update 2005; World Health Organization, 2005.
- 6 O. Yu, L. Sheppard, T. Lumley, J. Q. Koenig and G. G. Shapiro, Effects of ambient air pollution on

- symptoms of asthma in Seattle-area children enrolled in the CAMP study, *Environ. Health Perspect.*, 2000, **108**, 1209–1214.
- 7 T. F. Mar, T. V. Larson, R. A. Stier, C. Claiborn and J. Q. Koenig, An analysis of the association between respiratory symptoms in subjects with asthma and daily air pollution in Spokane, Washington, *Inhalation Toxicol.*, 2004, **16**, 809–815.
- 8 J. S. Lighty, J. M. Veranth and A. F. Sarofim, Combustion aerosols: factors governing their size and composition and implications to human health, *J. Air Waste Manage. Assoc.*, 2000, **50**, 1565–1618; discussion 1619–1622.
- 9 F. Laden, J. Schwartz, F. E. Speizer and D. W. Dockery, Reduction in fine particulate air pollution and mortality, *Am. J. Respir. Crit. Care Med.*, 2006, **173**, 667–672.
- 10 C. A. Pope III and D. W. Dockery, Health effects of fine particulate air pollution: lines that connect, *J. Air Waste Manage. Assoc.*, 2006, **56**, 709–742.
- 11 M. Jerrett and M. Finkelstein, Geographies of risk in studies linking chronic air pollution exposure to health outcomes, *J. Toxicol. Environ. Health, Part A*, 2005, **68**, 1207–1242.
- 12 M. Jerrett, R. T. Burnett, R. Ma, C. A. Pope, D. Krewski, K. B. Newbold, G. Thurston, Y. Shi, N. Finkelstein, E. E. Calle and M. J. Thun, Spatial Analysis of air pollution and mortality in Los Angeles, *Epidemiology*, 2005, **16**, 727–736.
- 13 E. Nethery, S. E. Leckie, K. Teschke and M. Brauer, From measures to models: an evaluation of air pollution exposure assessment for epidemiological studies of pregnant women, *Occup. Environ. Med.*, 2008, **65**, 579–586.
- 14 R. Morello-Frosch, M. Pastor and J. Sadd, Environmental Justice and Southern California's "Riskscape", *Urban Affairs Review*, 2001, **36**, 551–578.
- 15 Z. Chowdhury, R. D. Edwards, M. Johnson, K. Naumoff Shields, T. Allen, E. Canuz and K. R. Smith, An inexpensive light-scattering particle monitor: field validation, *J. Environ. Monit.*, 2007, **9**, 1099.
- 16 R. L. Unger, Particle sensor and related method offering improved particle discrimination 1999.
- 17 J. Kasumba, P. K. Hopke, D. C. Chalupa and M. J. Utell, Comparison of sources of submicron particle number concentrations measured at two sites in Rochester, NY, *Sci. Total Environ.*, 2009, **407**, 5071–5084.
- 18 P. Kulkarni, P. A. Baron and K. Willeke, *Aerosol Measurement: Principles, Techniques, and Applications*, John Wiley & Sons, 2011.
- 19 A. S. Wexler and S. L. Clegg, Atmospheric aerosol models for systems including the ions H^+ , NH_4^+ , Na^+ , SO_4^{2-} , NO_3^- , Cl^- , Br^- , and H_2O , *J. Geophys. Res.*, 2002, **107**, 14.