# A Review on Real-Time Aerosol Measurement Techniques and their Correlations

Sam Cox, Heejin Cho and Charles A. Waggoner
Institute for Clean Energy Technology, Mississippi State University
205 Research Blvd Starkville, MS 39759

## ABSTRACT

Measuring aerosol removal rate (i.e., filtering efficiency) in an accurate manner is the single most important aspect in air filter performance tests. Various aerosol measurement instruments can be used in filter tests to report aerosol concentrations over a range of particle size, each with their own classifying technique. When two or more instruments are used in filter tests and there is no agreement between measurements from different instruments, it is imperative to understand the fundamental principles of each instrument characterizing the physical properties of aerosols and to apply a systematic approach to correlate measurements. This study intends to provide a review of different particle sizing techniques, to expand how they are used to measure aerosol concentrations, and to review correlation between different instruments. Different aerosol measurement techniques used in real-time instruments, such as Scanning Mobility Particle Sizer (SMPS), Aerodynamic Particle Sizer (APS), and Laser Aerosol Spectrometer (LAS), and correlation models for those instruments available in literature are first reviewed. Each instrument relies on different particle properties such as size, shape, refractive index, and density. With the different measuring techniques, different size ranges are measured each with an overlapping region to other instruments. Real-time measurements of various types of aerosols are made using the aforementioned instruments in a full-scale test stand for radial flow filters tested in the Institute for Clean Energy Technologies (ICET) at Mississippi State University (MSU). The effectiveness of correlation models found in literature are tested with those measurements. In addition, research gaps with respect to correlation among measurements from different instruments are discussed here.

## INTRODUCTION

A variety of different instruments can be used to measure aerosol concentrations over a range of particle size. They can broadly be placed into two different groups: gravimetric (i.e. Pilat impactors and filter based measurements), and real time instruments (i.e. scanning mobility particle sizer (SMPS), laser aerosol spectrometer (LAS), aerodynamic particle sizer (APS), etc.). These instruments use different characteristic particle sizes to be able to measure the aerosol concentration. For example, the APS measures the aerodynamic diameter of a particle by accelerating it through a nozzle and measuring the difference between the air velocity and particle velocity. This type of measurement bases the particle diameter on a reference particle, a sphere with a unit density. Therefore, density and shape cannot be found from these measurements, while being largely dependent on these factors. The SMPS uses a different strategy, which uses its electrical mobility with assumed bulk aerosol properties. Due to the lack of unknowns and assumptions in these measurement models, some particle characteristics remain unknown. By being able to analyze measurements from different devices, some important characteristics can be estimated that gives further insight into the effective aerosol properties [1].

## AEROSOL MEASURING INSTRUMENTS

For this review, the instruments under consideration are the SMPS, APS, and LAS. These instruments are actively used in HEPA filter testing at the Institute for Clean Energy Technology (ICET) at Mississippi State University. Table 1 shows the particle size range and the type of particle diameter being measured. A brief review of these instruments is provided.

Table 1. Select Aerosol Measuring Instruments and Size Ranges.

| Instrument | Particle Size Range (μm) | Particle Diameter Measured |
|---|---|---|
| APS | 0.5 – 20 | Aerodynamic Diameter |
| LAS | 0.09 – 7.5 | Geometric Diameter |
| SMPS | 0.025 – 1* | Electronic Mobility Diameter |

* Dependent on Differential Mobility Analyzer (DMA) and software.

The APS measures the aerodynamic diameter of the particles being sampled by accelerating the sampling stream through a nozzle and measuring the velocity of the particles with laser gates. The difference in velocity of the particle and the air stream can be directly correlated to the settling time of a spherical particle of unit density. The aerodynamic diameter and physical diameter is equal for a sphere of unit density. This is only possible by (1) diluting the sampling stream to not overflow the laser gates and (2) measuring particles with a high enough refractive index to be measured by laser gates. The smallest size of aerosol that can be measured can range from 0.2 to 0.5 μm for this type of measurement due to the optical detection. APS software has the option to produce mass distribution by assuming a bulk aerosol density [1-3]. Calibration of the APS is usually done with laboratory generated aerosol. These aerosols are spherical with a unit density. A calibration curve can be obtained to measure aerodynamic diameter as a function of the difference in velocity between the air stream and particles. Different methods of calibration exist explained by Baron [4].

The LAS correlates the light scattering intensity of a particle to its geometric size. The LAS differs from other optical based measurements by collecting the wide angle scatter of the particles, which greatly improves the correlation accuracy, producing a monotone calibration curve. This instrument also requires dilution of the sampling stream as to prevent coincidence counting. Because of this increased accuracy, larger size ranges and greater resolution can be achieved. The LAS offers the widest particle size range of all instruments in this paper [5]. The counting efficiency of laser aerosol instruments can drop dramatically around 150 nm. Lawless et al. [6] propose a stochastic reconstruction algorithm applied post data collection to improve the accuracy of the collected data.

The SMPS relies on a Differential Mobility Analyzer (DMA) which classifies particles based on their electronic mobility (i.e. selects a certain mobility diameter to measure). This is coupled with a condensation particle counter (CPC) to count the classified particles. A DMA is considered a first order measurement because of its basis on physical principals rather than correlation models to produce the particle size. The SMPS and other DMA based measuring instruments are considered the most accurate for small size ranges (8 nm to 200 nm). The SMPS particle concentration can be converted to mass concentration by assuming a bulk density of the aerosols that applies to all size ranges. The SMPS is commonly coupled with an APS to produce a large size distributions of aerosol samples [7-9].

## INSTRUMENT CORRELATION STUDIES

A limited number of studies have investigated the benefits of correlating different aerosol instruments. Valuable aerosol properties can be determined from comparing different instruments such as shape factors, particle densities, etc. In addition to determining properties that can potentially effect filter performance, different size range distributions from different instruments can be combined into a single size distribution. This section presents case studies which present methods of instrument correlation studies.

DeCarlo et al. [10] provides a comprehensive overview of base equations and theory for measuring aerosol distributions. Part 2 of this study combines aerodynamic and electronic mobility diameters distributions to estimate the composition, shape factor, size, and fractal dimension of particles as a function of fuel equivalence ratio. A system of equations is formulated to be able to solve for these factors by an iterative method. To reduce the system of equations, the particles were assumed to have no internal voids (thereby adding uncertainties in density) and the dynamic shape factor was assumed to be equal in all flow regimes. Despite the estimations, the particle mass can still be estimated within 10%. It is important to note that this study was performed with suit from combustion and these assumptions may not provide the same accuracy under different types of aerosols [11]. Khlystov et al. [12] performed a similar study and propose an algorithm to combine electrical mobility and aerodynamic size distribution data. The algorithm focuses on finding a size correction factor to minimize the error of power law fits of the SMPS and APS. It is a straightforward procedure but assumes the size correction factor is constant in the overlapping size ranges. This is assumed to be a valid assumption given the small overlapping size range [12]. This method improves the comparison of the combined SMPS-APS size distribution with other instruments and increases the accuracy of estimated material density and dynamic shape factor. Khlystov et al. study [12] was influenced by another study conducted by Shen et al. [8] in which the SMPS-APS system is evaluated against a DataRam nephelometer and a Micro-Orifice Uniform Deposit Impactor (MOUDI). The study showed that the SMPS-APS system also showed good agreement with the MOUDI, but the quality of correlation was dependent on the relative size range. The MOUDI and SMPS-APS combination is also used in Khlystov et al. [12], and quality of fit is greatly improved when the proposed algorithm is used to combine the SMPS-APS into one size distribution based on electrical mobility diameter. Some assumptions still need to be made to allow this method to work such as dynamic shape factor and particle density in each size range. It can be hard to decipher this information without other instruments available.

Aerosol density is one of the most important measurements as it allows conversion from number concentration to mass concentration. In this case of filter testing, a single material density is often assumed for the challenge aerosol and iteratively adjusted to match mass loading curves to measured mass loaded post testing. If density per size bin could be estimated, more thorough analysis could be conducted. Mcmurry et al. [14] conducted a study to find the effective density of a particle size range using a tandem DMA setup in conjunction with an aerosol particle mass analyzer and CPC. The APM uses centripetal force (electronic mobility) to find the particle mass by spinning the particle until the inertia and centripetal forces are equal. Essentially, the DMA is used to classify the particles based on electronic mobility diameter and the APM measuring the mass distribution for the measured particles. More detailed information can be found from Ehara et al [13]. By knowing the mass and the electrical mobility diameter (assuming a spherical shape) an effective density can be estimated [14]. An APM is not available at the ICET, therefore this method cannot be tested later on. Hand et al. [15] also propose a method for retrieving effective density based on particle size distribution from three different instruments: DMA, Optical Particle Counter (OPC), and APS. It also relies on the overlapping regions between the OPC-DMA measurements and the OPC-APS measurements. An algorithm is introduced which can estimate the refractive index (not important for filter testing) and the effective density of aerosols per size bin. An OPC is also not currently available at the ICET. This method is heavily dependent on the Twomey algorithm to invert distribution data before attempting to fit the overlapping regions [15].

## CASE STUDY

This sections intends to use measurements obtained from different aerosol instruments upstream during filter testing to test the review of correlation studies. Some instruments used in the correlation studies are not currently available at the ICET or were not used during filter testing, notably the OPC and APM. Most filter testing uses a combination of SMPS, APS, LAS, Mark V Pilat Impactor, and ELPI. The SMPS, APS, and LAS are used for active analysis while the Pilat Impactor are generally used for verification.

The method of combining SMPS and APS data by Khlystov et al. [12] is used on representative test aerosol from a filter test performed at the ICET. LAS data is also compared to the two size distributions in the following figures. During filter testing, Alumina is used as the challenge aerosol to measure differential pressure versus mass loading and filter efficiency at selected differential pressures. All instruments iso-kinetically sample the main air stream and a dilution ratio of 20 and 1500 are applied to the APS and LAS respectively. The concentrations of the SMPS, APS and SMPS are shown in Figure 1. Khlystov et al. use a power law curve fit of the normalized aerosol distribution, but this study uses a log-normal distribution curve fit given by Eq. (1). Since the smallest 3 or 4 data points in the APS are generally unreliable due to the aerosol refractive index in that size range, they are omitted to enhance the fit of the log-normal curve [2].

$$\frac{df}{dlog(d_p)} = \frac{a}{\sqrt{2\pi}\log(\sigma_G)} \exp\left(-\frac{\log(d_p)-\log(CMD)^2}{2(\ln(\sigma_g))^2}\right) \tag{1}$$

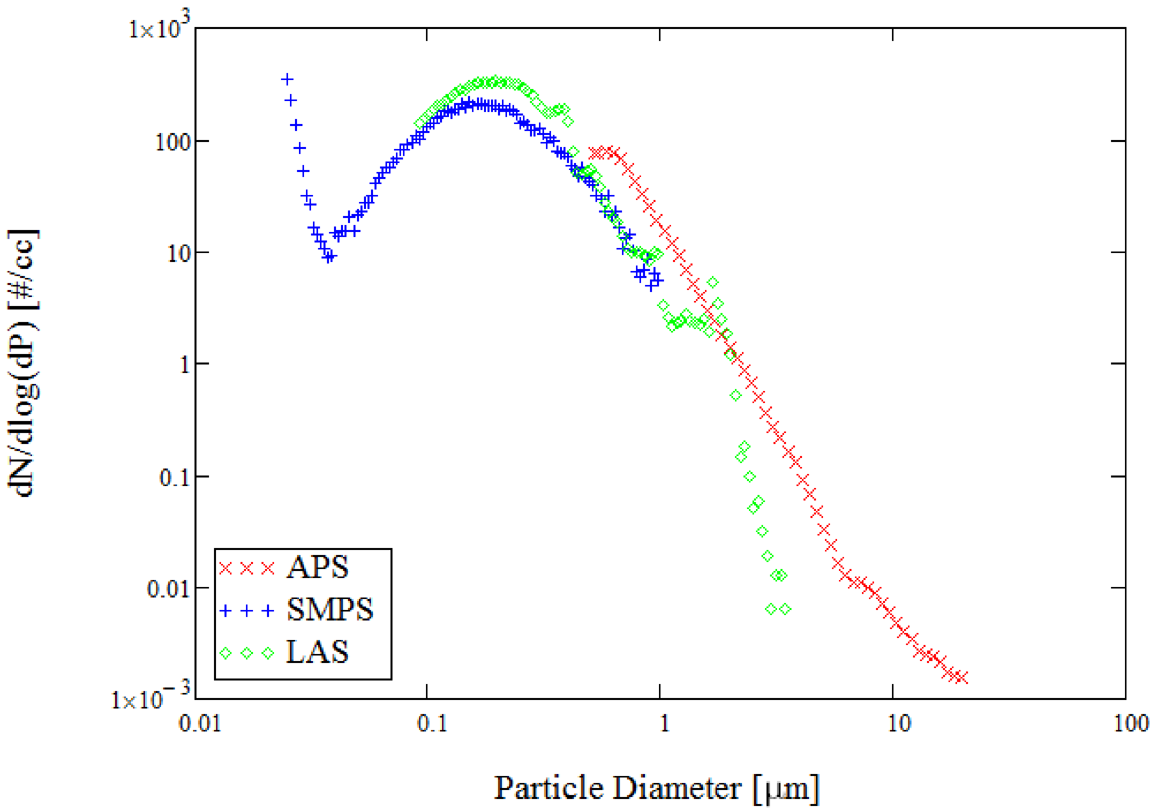


Figure 1. APS, SMPS, LAS Concentrations.

Next, the overlapping size range is determined upon for the SMPS and APS data. Khlystov et al. use 0.5 to 0.8 micrometers. The overlapping size range is considered static and is not updated between iterations. Khlystov et al. propose a cost function to minimize as a function of a size correction factor, given in Eq. (2).

$$S^2(c) = \frac{1}{n_2-n_1}\sum_{i=n1}^{n2}\left[\log\left(N_s(D_i)\right) - \log\left(N_a(D_i c)\right)\right]^2 \tag{2}$$

where $n_2$ and $n_1$ are the first and last size bin of the overlapping size range, $N_s$ and $N_a$ are the number concentration as a function of particle diameter, $D_i$, of the SMPS and APS respectively, and $c$ is the correction factor. This size correction factor reduces or increases the aerodynamic particle diameter given by the APS, effectively shifting the APS data left or right. This size correction factor allows the aerodynamic diameter to be approximately converted to electronic mobility diameter or vice versa. For alumina aerosol on one test, the size correction factor is 1.6, meaning the aerodynamic diameter is effectively 1.6 times greater than the electronic mobility diameter in the overlapping size range. Figure 2 compares the updated APS data and the SMPS data. While the method shown approximately converts the aerodynamic diameter into the electronic mobility diameter, the inverse can be achieved if aerodynamic diameter is more important, as in medicine, human health, or filter testing.

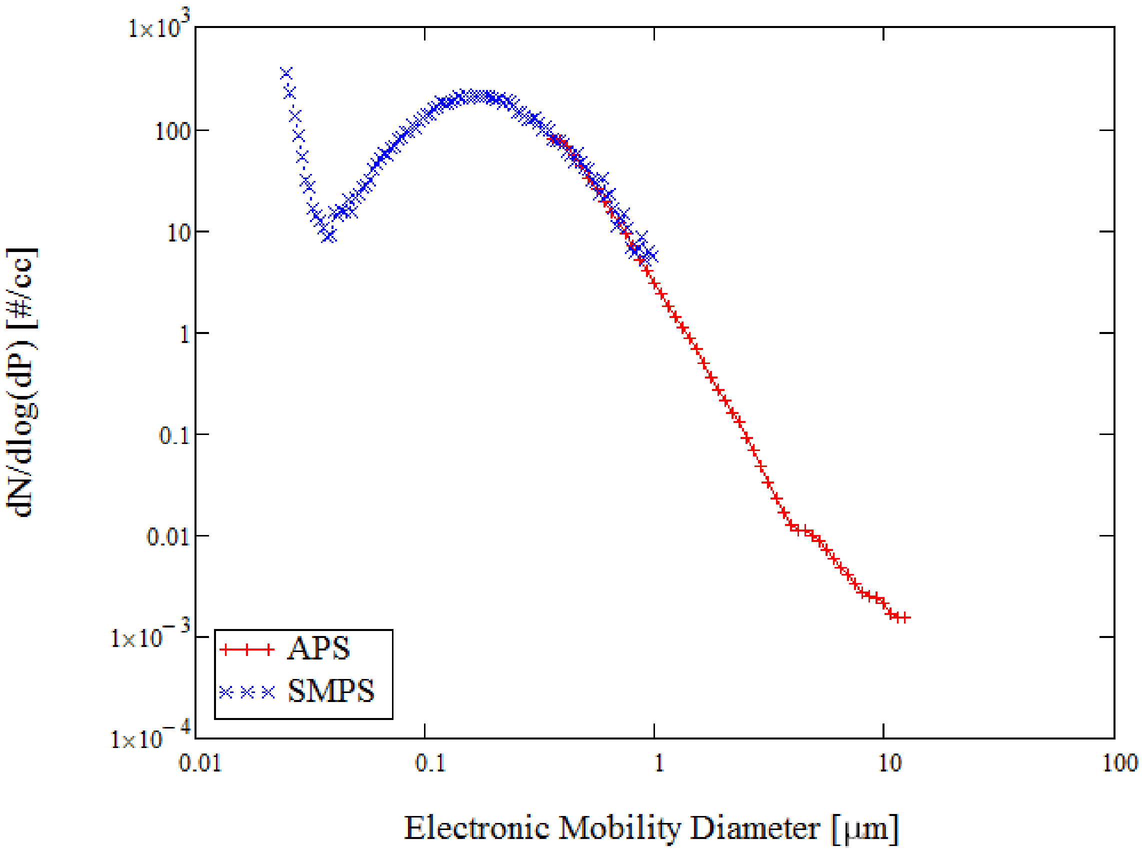


Figure 2. SMPS and Converted APS Concentrations.

In this case a simplified equation for the relationship between electronic mobility and aerodynamic diameter is given in Eq. (3) [10]. Assuming this relationship holds true, the size correction found by the algorithm is the square root of the ratio between the effective shape diameter and effective particle density in the overlapping size range given the unity density is 1.

$$d_e = \frac{d_a}{\sqrt{\frac{\rho_p}{\rho_0 \chi}}} \tag{3}$$

where $D_e$ is the electrical mobility diameter, $D_a$ is the aerodynamic diameter, $\rho_p$ is the particle density, $\rho_0$ is unit density, and $\chi$ is the dynamic shape factor. By rearranging Eq. (3) into Eq. (4) and assuming a shape factor of 1, the effective density of the particles in the overlapping size range can be found by Eq. (5).

$$\left(\frac{d_a}{d_e}\right)^2 = \frac{\rho_p}{\chi} \tag{4}$$

By replacing the ratio of the aerodynamic and electrical mobility diameter with $c$ found from the algorithm and considering an effective density instead a particle density, Eq. (5) is derived.

$$\rho_{eff} = \chi\, c^2 \tag{5}$$

Where $\rho_{eff}$ is the effective density for the particles in the overlapping size range. In this case, alumina is used as the testing aerosol and by this method, an effective density of 2.60 g/ccm is estimated from the algorithm where the material density for alumina is 2.42 g/ccm. There are other definitions of effective density mentioned in literature outlined by DeCarlo et al. [s10]. A common one that only relies on electronic mobility diameter and vacuum aerodynamic diameter is proposed by Jimenez et al. [16]. The relationship between the mobility diameter and vacuum aerodynamic diameter is not dependent upon the square root of the particle density divided by the dynamic shape factor and unit density. Therefore, this relationship does not apply for APS data and a second definition is derived.

Different methods exist that can more accurately estimate the effective density as described earlier such as Mcmurry et al. [14] who use a DMA/APM setup and Khlystov et al. [12] who use a MOUDI impactor and APS/SMPS. Inversely, given an effective density, an effective shape factor can be found from Eq. (5) as well. Care must be taken when applying these estimated parameters to the entire size distribution, as these values can vary throughout particle size.

## CONCLUSION

Aerosol measurement techniques are fundamental to proper filter testing. By utilizing a wide array of near real time aerosol measurements, important characteristics of the challenge aerosol can be found and the filtering response can be analyzed in greater detail. A series of correlation techniques among different aerosol instruments were reviewed and a case study was conducted based on SMPS and APS data during a nuclear grade HEPA filter test. It is found that the SMPS and APS correlation can be a useful tool in future filter testing. Most studies estimate bulk aerosol properties, but there is an opportunity to estimate these per size bin, although the uncertainty in these estimations will rise along with complexity. Correlation models using the Electrical Low Pressure Impactor (ELPI) and LAS have been understudied, but the LAS could potentially replace the OPC used in previous studies. By expanding on the reviewed articles and developing more correlation models, advanced algorithms can be developed to examine filter response based on effective aerosol properties.

## REFERENCES

[1] P. H. McMurry, “A review of atmospheric aerosol measurements,” *Atmos. Environ.*, vol. 34, no. 12–14, pp. 1959–1999, 2000.

[2] W. C. Hinds, *Aerosol technology: properties, behavior, and measurement of airborne particles*, 2nd ed. John Wiley & Sons, 1999.

[3] TSI Incorporated, “Aerodynamic Particle Sizer Model 3321”.

[4] P. Baron, "Calibration and Use of the Aerodynamic Particle Sizer (APS 3300) ," *Aerosol Sci. Technol.*, vol. 5, no. 1, pp. 55–67, 1986.

[5] TSI Incorporated, "Laser Aerosol Spectrometer Model 3340".

[6] P. A. Lawless and S. V. R. Mastrangelo, "Theoretical Response of Laser Aerosol Spectrometers and Data Inversion by Stochastic Reconstruction," *Aerosol Sci. Technol.*, vol. 38, no. 5, pp. 409–423, 2004.

[7] TSI Incorporated, "Scanning Mobility Particle Sizer SpectroMeter (SMPS) Model 3936".

[8] S. Shen, P. A. Jaques, Y. Zhu, M. D. Geller, and C. Sioutas, "Evaluation of the SMPS-APS system as a continuous monitor for measuring PM2.5, PM10 and coarse (PM2.5-10) concentrations," *Atmos. Environ.*, vol. 36, no. 24, pp. 3939–3950, 2002.

[9] R. Friehmelt, H. Buttner, and F. Ebert, "On-line Characterisation of Aerosols-Comparability and Combination of Selected Measuring Devices," *KONA Powder Part. J.*, vol. 18, no. 18, pp. 183–193, 2000.

[10] P. DeCarlo , J. Slowik, D. Worsnop, P. Davidovits, and J. Jimenez, "Particle Morphology and Density Characterization by Combined Mobility and Aerodynamic Diameter Measurements. Part 1: Theory," *Aerosol Sci. Technol.*, vol. 38, no. 12, pp. 1185–1205, 2004.

[11] J. Slowik, K. Stainken, P. Davidovits, L. R. Williams, J. T. Jayne, C. E. Kolb, D. Worsnop, Y. Rudich, P. DeCarlo, and J. Jimenez, "Particle Morphology and Density Characterization by Combined Mobility and Aerodynamic Diameter Measurements. Part 2: Application to Combustion-Generated Soot Aerosols as a Function of Fuel Equivalence Ratio," *Aerosol Sci. Technol.*, vol. 38, no. 12, pp. 1206–1222, 2004.

[12] A. Khlystov, C. Stanier, and S. N. Pandis, "An Algorithm for Combining Electrical Mobility and Aerodynamic Size Distributions Data when Measuring Ambient Aerosol Special Issue of Aerosol Science and Technology on Findings from the Fine Particulate Matter Supersites Program," *Aerosol Sci. Technol.*, vol. 36, no. 4, pp. 411–416, 2002.

[13] K. Ehara, C. Hagwood, and K. J. Coakley, "Novel method to classify aerosol particles according to their mass-to-charge ratio - Aerosol particle mass analyser," *J. Aerosol Sci.*, vol. 27, no. 2, pp. 217–234, 1996.

[14] P. H. McMurry, X. Wang, K. Park, and K. Ehara, "The Relationship between Mass and Mobility for Atmospheric Particles: A New Technique for Measuring Particle Density," *Aerosol Sci. Technol.*, vol. 36, no. 2, pp. 227–238, 2002.

[15] J. L. Hand and S. M. Kreidenweis, "A new method for retrieving particle refractive index and effective density from aerosol size distribution data," *Aerosol Sci. Technol.*, vol. 36, no. 10, pp. 1012–1026, 2002.

[16] J. L. Jimenez, R. Bahreini, D. R. Cocker, H. Zhuang, V. Varutbangkul, R. C. Flagan, J. H. Seinfeld, C. D. O'Dowd, and T. Hoffmann, "New particle formation from photooxidation of diiodomethane (CH2I2)," *J. Geophys. Res.*, vol. 108, no. D10, p. -, 2003.

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