Measurement of mass of aerosol particles

Ehara Kensei[1*], Charles Hagwood[2], Kevin J. Coakley[2], Fukushima Nobuhiko[3],
Kittichote Worachotekamjorn[4], Tajima Naoko[1] and Sakurai Hiromu[1]

An aerosol particle mass analyzer (APM) which classifies aerosol particles according to their mass has been developed. Mass distributions of aerosol particles are measured by the APM combined with a particle counting device. Particle masses can be measured in the range 3×10^{-18} g to 2×10^{-12} g, which partially fills the mass range that has not been covered by existing mass measuring instruments such as mass spectrometers and conventional balances. The invention of the APM has led to a variety of new techniques for evaluating aerosol particle properties such as effective density, material density, porosity, fractal dimension, and mass concentration of suspended particulates, among others. This article describes the principle of the APM, its features differentiating it from other instruments for classifying aerosol particles, and its applications to characterization of aerosol particles. The significance of measurement of particle mass in aerosol science, and the historical process that has led to commercialization of the APM are also described from the viewpoint of "synthesiology."

1 Introduction

A two-phase system consisting of suspended solid or liquid particles and the surrounding gas, typically air, is called an aerosol. Aerosols have attracted much attention from a variety of fields, because of their involvements in a broad range of phenomena such as potential damage to health and the environment by airborne nanoparticles, transport of radioactive elements through the air, global warming, and particle contamination in cleanrooms, among others. Recent interests in aerosols are due not only to these undesirable effects, but also to their possible useful applications such as aerosol processes for creation of novel materials.

In order to understand phenomena in which aerosol particles are involved in some way or other, measurement of various properties of the aerosol particles is needed.[1] Among these properties, particle size (particle diameter, if the particle in question is spherical) is an especially important quantity to be measured, because it significantly affects many of the aerosol-related phenomena. Aerosol particles are, however, often non-spherical, and a geometric diameter cannot be defined unequivocally for them; instead, several kinds of effective diameters are defined and measured. On the other hand, particle mass is a quantity inherent to each individual particle: it is uniquely defined even for a non-spherical particle. It is a direct measure of the amount of substance contained in an individual particle, and has crucial effects on particle motion and other physical phenomena; hence it should be no less important than the particle size. Until recently, however, there has been no established method for measuring mass of aerosol particles. In the present article, the aerosol particle mass analyzer (APM) that has enabled measurement of mass of aerosol particles[†] is described from the viewpoint of "synthesiology."

2 Significance of particle mass and its measurement

In this section, the relationship of particle mass with some other particle properties is described first in Subsection 2.1. Then the significance of knowing mass of aerosol particles in understanding their impacts on various human activities is discussed in Subsection 2.2. Finally in Subsection 2.3, how we became interested in their measurement is briefly reviewed.

2.1 Particle mass and equivalent diameters

The electron microscope images of particles in Fig. 1 show a variety of shapes particles can exhibit depending on how they are produced. Particle mass is a physical quantity that can be unambiguously defined for particles of any shapes, but particle size is not. For non-spherical particles, such as shown in Figs. 1(b) to 1(d), an equivalent diameter is used instead of the geometric diameter. An equivalent diameter is defined as the diameter of a spherical particle that has the same value of a certain physical quantity as the particle under consideration. Among representative equivalent diameters are: kinematic equivalent diameters including the mobility equivalent diameter and the aerodynamic diameter; geometric equivalent diameters including the volume equivalent diameter and the surface area equivalent diameter; and optical equivalent diameters including the light

1. National Institute of Advanced Industrial Science and Technology   1-1-1 Umezono, Tsukuba, 305-8560, Japan   *E-mail: ehara_k@meiji.ac.jp 2. National Institute of Standards and Technology, USA, 3. Kanomax Japan Inc.   2-1 Shimizu, Suita, 565-0805, Japan, 4. Prince of Songkla University, Thailand

scattering equivalent diameter.[1] In the following, kinematic equivalent diameters are described in some detail, because they are closely related to the particle mass.

Mechanical mobility†† is the ratio of the velocity of a particle in motion under the influence of a certain external force to that force. Mobility equivalent diameter, D_B, is defined as the diameter of a spherical particle having the same mechanical mobility, B, as the particle in question. The mechanical mobility of a spherical particle with diameter D can be expressed as

$$B = \frac{C(D)}{3\pi\eta D},$$ (1)

where η is the viscosity of the surrounding air, and $C(D)$ is the slip correction coefficient (a numerical factor expressing the deviation of drag force experienced by a particle from Stokes' law). The mobility equivalent diameter, D_B, of a non-spherical particle with mobility B is given by solving equation (1) for D. Because the drag force depends only on the spatial extension of the particle and not on its mass nor density, so do D_B and B. The product qB of a particle of charge q and mechanical mobility B is known as electrical mobility ($z = qB$), and it represents the velocity of a charged particle in an electrostatic field of a unit strength. Because methods to measure electrical mobility are well developed compared to methods to measure mechanical mobility (see Subsection 3.2), the electrical mobility is often measured in place of the mechanical mobility. In the following, the term "mobility" is used for simplicity to indicate either the electrical mobility or the mechanical mobility, as far as there is no fear of confusion.

Aerodynamic diameter D_A is another kinematic equivalent diameter; it is the diameter of a standard-density ($\rho_0 = 1$ g/ cm³) sphere (i.e., a spherical water droplet) which has the same terminal velocity as the particle in question when it settles under gravity. The mass m of a spherical water droplet with diameter D is $\rho_0\pi D^3/6$, and its terminal settling velocity v_T is given by mBg. (g is the gravitational acceleration). Hence the product mB in this expression can be rewritten as

$$mB = \frac{C(D)D^2\rho_0}{18\eta}.$$ (2)

If we measure v_T in the gravitational field (or in some acceleration field of a known magnitude), the quantity mB can be determined as v_T/g. Once the value of mB is known in this way, the aerodynamic diameter, D_A, is obtained by solving equation (2) for D regardless of its particle shape. Particles with various shapes migrate at the same terminal velocity in a given acceleration field, as far as they have the same value of D_A. This is the major reason why we want to know the value of D_A.

Equations (1) and (2) indicate that if we know two of the three quantities, m, D_B, and D_A, the rest can be known from them. For example, the value of D_A of a particle with a known m can be derived from a measurement of D_B, and vice versa. Recent studies show that, in general, if simultaneous measurement of particle mass m and some other particle property is conducted, various properties of the particle can be derived from them; a variety of such examples are given in Section 4.

2.2 Endpoint effects of aerosols
We denote an impact that aerosol particles ultimately have on human activities, whether desirable or not, as an endpoint effect. To control an endpoint effect of aerosols, it is crucial to evaluate its magnitude. However, it is often not easy nor useful to directly evaluate the endpoint effects of aerosols.
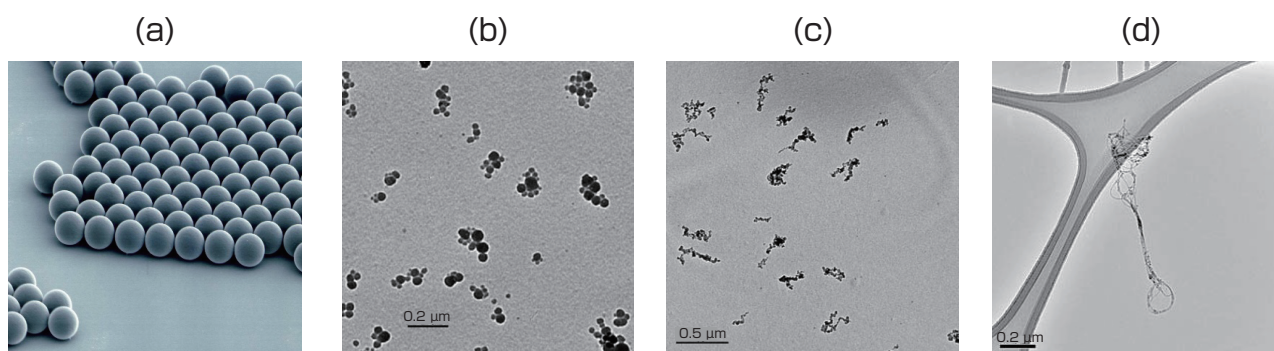
(a) (b) (c) (d)

Fig. 1 Examples of the shape of various particle types: (a) polystyrene latex particles, (b) Al₂O₃ particles, (c) diesel exhaust particles, (d) single-walled carbon nanotubes. Photograph (a) is by courtesy of JSR corporation. Photographs (b) and (c) are reproduced from reference [2], and photograph [d] from reference [3], both with permission.

† Though we use the phrase "measurement of mass" in this article for simplicity, it would be more adequate to use the phrase "measurement of mass distribution," because in aerosol measurements, we are almost always interested in a property of the particles contained in an aerosol as a whole, and not of one specific particle. Also, note that "mass of aerosol particles" here does not imply the total mass of particles collected, for example, on a particulate filter, but implies mass of individual aerosol particles.
†† The particle mobility B is in general a tensor, but is treated here as a scalar for simplicity.

– 94 –

Table 1. Forces employed and particle properties classified in representative instruments for classifying aerosol particles.

Type of force / Instrument / Quantity involved	Electrostatic Charge (q)	Diffusion [note 1]	Inertial Mass (m)	Centrifugal Mass (m)	Gravitational Mass (m)	Drag Mobility (B)	Property to be classified
Differential mobility analyzer	✓					✓	Mobility equivalent diameter
Diffusion battery		✓				✓	Mobility equivalent diameter
Impactor			✓			✓	Aerodynamic diameter
Centrifugal classifier [note 2]				✓		✓	Aerodynamic diameter
Elutriator					✓	✓	Aerodynamic diameter

[note 1] The diffusion force is expressed as $-kT(\nabla n)/n$, where n is the particle number concentration, k the Boltzmann constant, and T the thermodynamic temperature, and does not depend on any properties of individual particles.

[note 2] Examples of centrifugal classifiers include a Stöber centrifuge, a Goets spectrometer, and a cylindrical aerosol spectrometer.

For example, in the case of potential health hazards of nanoparticles that has attracted recent concern with respect to the emerging nanotechnology, the endpoint effect would be a long-term deterioration of health of people who have inhaled the nanoparticles. However, it is impossible to conduct an experiment to quantify such an effect with humans as experimental objects. Moreover, for the purpose of preventing the hazardous effect, evaluation of the endpoint effect after a person's health is damaged is useless. What we should do instead is to first characterize the nanoparticles with respect to their various physical and chemical properties, and then clarify the relationship of each particle property with biological toxicity of the particles through, for example, inhalation studies using experimental animals or desirably through *in vitro* analyses. Control of an endpoint effect of aerosols will be only possible on the basis of our knowledge of such relationships, combined with measurements of relevant properties of the particles concerned.

In general, it is *a priori* unknown which particle property is relevant to a given endpoint effect of an aerosol. For example, before we conduct inhalation exposure tests, we do not know which property of nanoparticles is involved in their suspected biological toxicity; size,[†] mass, shape, surface area, chemical component, or some other property. Hence, many methods for measuring as many kinds of particle properties as possible should be available when quantification of the endpoint effect of some specific aerosol is required. In particular, methods to measure the mass of aerosol particles are essential.

2.3 Motivation for starting research on particle mass measurement

Before our research and development efforts commenced, there was no practical method to measure aerosol particle mass. Further, the research community had not vigorously promoted the development of such a method. Hence, primarily on our own initiative, we developed a new fundamental measurement method for aerosol science and technology. From the beginning, we anticipated that such a method would enable new particle characterization techniques since mass is a very basic quantity. In our view, this expectation has been realized as described later in Section 4.

3 Aerosol particle mass analyzer (APM)

3.1 Existing particle classification instruments

A typical method for measuring the distribution of a specific particle property is to use an instrument that can extract only particles with property values in a narrow interval centered on an adjustable central value. The distribution of the property can be obtained by quantifying the extracted particles for various values of the property. Such quantification can be done, for example, with a single-particle counting device such as a condensation particle counter (CPC) or a light scattering airborne particle counter (LSAPC); an aerosol electrometer, which detects the electric current carried by charged particles to determine the particle number concentration; or a microbalance to weigh particles collected onto a particulate filter.

Table 1 shows the forces employed and the particle properties classified by representative existing instruments for classification of aerosol particles.[4] When a particle moves at velocity v relative to the surrounding air, it experiences a drag force equal to $-v/B$. We can regard particle classification by the instruments listed in Table 1 as being realized by balancing a

---

† The term "size" is used in this article to indicate a rough measure of the spatial extent of a generally non-spherical particle.

specific force externally applied to the particle with the drag force. Take the differential mobility analyzer as an example. A particle with charge q placed between electrodes experiences an electrostatic force, and migrates at a certain velocity. The condition that the electrostatic force (proportional to q) equals the drag force (inversely proportional to B) is satisfied at a specific value of electrical mobility qB. Only particles whose electrical mobility is in a narrow interval about this value are separated out (see Subsection 3.2). Similarly, for instruments that employ a force proportional to particle mass m, particle classification is realized by balancing that force with the drag force. For this type of instrument, the particle property to be classified turns out to be the product mB, or equivalently the aerodynamic diameter.

Thus the drag force, being dependent on particle mobility, plays an essential role in the existing instruments for classification of aerosol particles. As a result, the property according to which particles are classified inevitably has a dependence on mobility, and thus classification according to particle mass cannot be realized. In the case of the APM, the drag force does not play an essential role, which enables classification by mass. Further details of the APM is described in the following.

3.2 Principle of the APM

In this subsection, the operating principle of the APM is described while contrasting it to that of the differential mobility analyzer (DMA).[5] The main part of the DMA consists of coaxial cylindrical electrodes as shown in Fig. 2. A sample aerosol is introduced through the slit at the upper
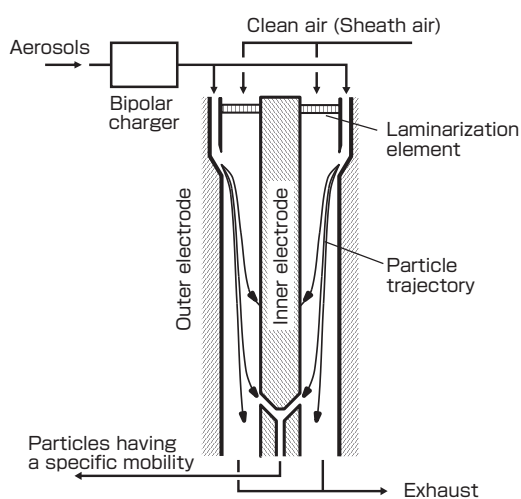
part of the outer electrode into clean air flowing parallel to the electrode axis. A charged particle in the aerosol migrates between the electrodes along a trajectory that depends on its electrical mobility, and only those particles having a certain electrical mobility are extracted through the slit at the lower part of the inner electrode. Using a CPC to count the particles exiting the DMA, we can measure the distribution of mobility equivalent diameter of the particles. The combined DMA-CPC system is extensively used in this way for the purpose of particle-size distribution measurement.

The APM similarly consists of coaxial cylindrical electrodes, but is constructed so that the electrodes, both the inner and outer ones, rotate at the same angular velocity (see Fig. 3). A charged particle introduced into the gap of the rotating electrodes migrates under the influence of an inward electrostatic force and an outward centrifugal force. These two forces balance when the particle has a specific mass-to-charge ratio. When the forces on a particle achieve this balance, the particle is transmitted through the electrode gap.[†] Because mass-to-charge ratio is the property to be classified, this instrument was termed aerosol particle mass analyzer.[6]

The existing classification instruments such as listed in Table 1 employ only one type of external force (note that drag force is not an external force), and make use of particle motion caused by that force. The operating principles of these instruments can be categorized as the so-called deflection method, in which difference in travel distance between particles having different values of the property is utilized for classification of that property. On the other hand, the APM employs two forces, electrostatic and centrifugal, which allows particles having a specific mass-to-ratio not to migrate relative to the surrounding air.[††] This feature of the
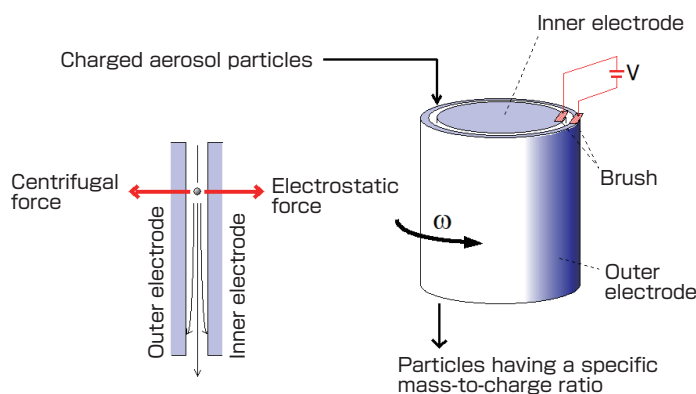

Fig. 2 Principle of the differential mobility analyzer (DMA).

Fig. 3 Principle of the aerosol particle mass analyzer (APM).

---

† For simplicity, the APM is treated in this article as a classifier of particle mass, though strictly it is a classifier of mass-to-charge ratio. We can often set up a measurement system so that particle charge is specified, and in such cases the particle mass can be readily inferred from the mass-to-charge ratio.

†† To be strict, particles that penetrate the electrode gap may have migrated in the radial direction of the cylindrical electrodes over a distance equals to the gap length at the maximum. Such particle migration in the radial direction determines the resolution in mass classification, but will not be discussed in this article.

APM makes mass classification possible. The principle of the APM is referred to as the so-called zero method, where a force balance reduces particle motion to zero.

The classification performance of the APM is characterized by the APM transfer function[†] $\Omega(m; V)$.[6] The transfer function is the ratio of an exiting number flux to a penetrating (incident) number flux of particles having a specific value of mass m. It depends on the voltage applied between the electrodes V, and the angular velocity of the electrodes ω. The symbol $\Omega(m; V)$ indicates that it is a function of m with V being a parameter (the ω dependence is not explicitly indicated for simplicity). The transfer function can be theoretically calculated by solving the equation of motion of particles within the electrode gap.

When aerosol particles with a mass distribution $f(m)$, where $f(m)\mathrm{d}m$ represents the number concentration of particles within a range $(m, m+\mathrm{d}m)$, are drawn to the APM operated at voltage V, the number concentration of particles exiting the APM is given by[††]

$$n(V)=\int_0^\infty f(m)\Omega(m;V)\mathrm{d}m. \qquad (3)$$

The concentration $n(V)$ normalized by the concentration of the particles entering the APM ($\int_0^\infty f(m)\mathrm{d}m$) defines the particle penetration rate. The concentration $n(V)$, or the particle penetration rate, as a function of V is called an APM spectrum. Based on measurements of $n(V_i)$ at various values of the applied voltage V_i, and a theoretical model for the mass distribution $f(m)$, one can estimate the parameters of the theoretical model by the method of least squares fitting.[7]

3.3 Studies on the APM by other groups
Since we publicized the principle of the APM, studies on the performance of the APM as well as on analysis of APM data have been conducted by several research groups. These studies include experimental evaluation[8] and theoretical analysis[9] of the APM transfer function, investigation of the effects of Brownian diffusion on the APM performance,[10]

and handling of the inverse problem for reconstruction of particle mass distributions.[11] We will not go into the details of these studies.

As already described, the APM was designed so that the inner and outer electrodes rotate at the same angular velocity. As a result of this feature, the point of equilibrium between the electrostatic and centrifugal forces for a particle having a given mass-to-charge ratio corresponds to a slightly unstable equilibrium, meaning that the potential energy experienced by the particle is the maximum, not minimum, at the equilibrium point. Olfert and Collings, then at Cambridge University, developed an instrument that they termed a Couette centrifugal particle mass analyzer (CPMA).[12][13] This instrument is similar in design to the APM except that the inner electrode rotates faster than the outer electrode, so that the point of equilibrium between the two forces corresponds to the minimum of the potential energy. Due to this feature, if the APM and CPMA are operated under conditions realizing the same resolution in classification, the CPMA can extract, in theory, more particles than the APM does for a given number flux of incident particles. This property is often advantageous in experiment, because quantification of extracted particles is easier when more particles are available. Unfortunately, however, probably due to difficulty in establishing the fluid-dynamically ideal Couette flow[†††] between the coaxial cylindrical electrodes, a CPMA that can achieve a theoretically expected performance has not yet been realized.[13] In what follows, the CPMA is not discriminated from the APM, and treated as one type of the APM.

3.4 The APM as an instrument for mass measurement
Figure 4 shows mass measurement ranges covered by representative instruments on a log-scale. The smallest change in mass detectable by high-resolution microbalances is about 0.1 µg. A tapered element oscillating microbalances (TEOM) collects a small amount of particulate matters suspended in the air such as PM2.5 on an oscillating element, and determines its mass from the change in the oscillation frequency. The TEOM covers the mass range of
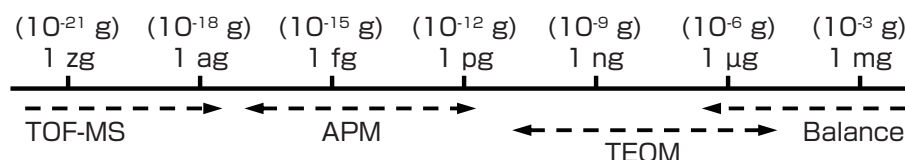

Fig. 4 Mass ranges covered by representative mass measuring instruments (TOF-MS: time of flight mass spectrometer, TEOM: tapered element oscillating microbalance).

[†] The term "transfer function" is adopted after the similar function that has been used extensively in analyses of DMA data.

[††] It is assumed here that all particles that enter the APM carry a single unit of charge. Such a condition can be approximately realized by installing a DMA upstream of the APM.

[†††] Couette flow is a flow of a viscous fluid between two surfaces, such as two flat plates or two cylindrical pipes, that have a finite relative velocity in their tangential directions. The components of the flow velocity parallel to the surfaces have a gradient perpendicular to the surfaces in the Couette flow.

approximately 10^{-11} g to 10^{-5} g.[14] On the other hand, mass of an atom or a molecule is measured by mass spectrometers, and a time-of-flight mass spectrometer (TOF-MS) among other types of mass spectrometers has a relatively broad measurement range, covering approximately 1.7×10^{-24} g to 1.7×10^{-18} g (1 D_a to 10^6 D_a).

The mass range of approximately 10^{-18} g to 10^{-11} g is not covered by existing instruments. The APM can partly fill this blank region, covering approximately 3×10^{-18} g to 2×10^{-12} g.[7] It should be noted, however, that the mode of measurement is different between the instruments. While the balance can measure the mass of a given object, the TEOM measures the mass of collected particles as a whole, and the APM and TOF-MS measure the mass distribution of a collection of particles.

4 Applications of the APM

Since the APM became available for practical use, several new methods of characterization of aerosol particles have emerged. Because they are considered direct outcomes of the invention of the APM, we describe them in some detail in the following.

4.1 Effective density[15]–[20]
The distribution of effective density of aerosol particles can be measured by connecting a DMA and an APM in series, and counting the number of separated particles by a CPC. The effective density is defined here as the mass of a particle, m, divided by the volume of a spherical particle, $\pi D_B^3/6$, that has the same mobility equivalent diameter D_B as the particle under consideration. If the particle is spherical, the effective density reduces to the particle density. Because the effective density reflects constituent materials and morphology of the particle, it carries information not available only through measurement of the particle size. The method of measuring the effective density using a combined DMA-APM system was first proposed by P. H. McMurry of the University of Minnesota and his colleagues.[15] Using this method in the city of Atlanta, they showed for the first time that airborne particles in urban areas are sometimes composed of two rather distinct components, one with a relatively large effective density and the other with a small effective density. Since the publication of their study, measurements of effective density for atmospheric particles in various places, as well as for various types of particles generated in laboratories have been conducted by a significant number of groups.[16]–[20]

4.2 Material density and particle density[2][21][22]
The density of an aerosol particle can be determined from its mass and volume; the former obtained with the APM, and the latter by some other means, typically by electron microscopy.[2] The density determined in this way is the so-called particle density, if the volume used in this procedure includes that of the voids within the particle, while it is the material density,

if not. Using this method, S. H. Kim and his colleagues of the University of Maryland succeeded in determining the density of multi-walled carbon nanotubes; in this case, the particle density including contribution from the hollow of the nanotubes.[21] By similar methods, the densities of diesel exhaust particles, metal particles, and metal oxide particles among other types of particles have been measured by several groups.[2][22]

4.3 Mass and volume[23]–[32]
The mass fraction of volatile materials contained in particles can be determined from the change in the mass upon heating the particles. Sakurai H., then at the University of Minnesota, and his colleagues used this technique to study the size dependence of the mass fraction of volatile materials in diesel exhaust particles, where an APM combined with a tandem DMA system (two DMAs connected in series) was utilized.[23]

When the material density of a particle is known, the volume of the particle can be determined from its mass. A. A. Lall of the University of California, Los Angeles, and his colleagues used an APM to experimentally determine the mass and volume of aggregated particles for the purpose of testing the validity of their "idealized aggregate theory," which is to predict the volume and number of primary particles in an aggregate particle from its mobility and the diameter of primary particles constituting it.[25]

Moteki N. and Kondo Y. of the University of Tokyo used an APM in their performance evaluation study of the single particle soot photometer (SP2), which uses the laser induced incandescence technique to measure the mass and mixing state of individual elementary-carbon particles in the atmosphere, which are considered to have significant impacts on global warming.[24]

4.4 Mass concentration[33][34]
The mass concentration of atmospheric particles with aerodynamic diameter smaller than a certain stipulated threshold (i.e., the total mass of such particles per unit volume of air) is an indicator of particulate pollution of the atmosphere. Well-known indicators include PM2.5, with the threshold equal to 2.5 μm, and suspended particulate matter (SPM), with the threshold equal to 10 μm. The reference method to measure the mass concentration is based on gravimetric measurement of the mass of the particulate matter collected onto an air filter. This method is considered accurate overall, but it also has drawbacks such that evaporation of volatile components and/or adsorption of foreign substances may occur during the collection process, impairing the reliability of measurement, and that a long collection time is often needed to collect enough amount of particles that can be measured with a desired accuracy.

K. Park of the University of Minnesota and his colleagues proposed an alternative way of measuring the mass

concentration of atmospheric particles.[33] In their method, the mean mass $\bar{m}(D_B)$ of the particles having a specific value of mobility equivalent diameter D_B is determined prior to the measurement by means of a combined DMA-APM-CPC system. The measurement is conducted for the number distribution of mobility equivalent diameter dN/dD_B using a combined DMA-CPC system. The mass concentration M can be obtained from calculating

$$M = \int \bar{m}(D_B)\frac{dN}{dD_B}dD_B.\qquad(4)$$

This method has an advantage that no measurement bias associated with the evaporation/adsorption phenomena during the particle collection process occurs, because neither measurement of $\bar{m}(D_B)$ nor that of dN/dD_B requires particle collection. Another advantage is that as far as the mean particle mass as a function of the mobility equivalent diameter, $\bar{m}(D_B)$, remains unchanged, the mass concentration can be determined solely from dN/dD_B, resulting in a relatively short measurement time even when the particle number concentration is significantly low.

4.5 Fractal dimension[35]–[49]
Aggregate particles, such as shown in Figs. 1(b) and 1(c), grow through coagulation of relatively small primary particles. It is known[50] that under certain conditions, the growth of such particles can be described by the following scaling law between the particle mass m and the mobility equivalent diameter D_B,

$$m \propto D_B{}^{d_\mathrm{f}}.\qquad(5)$$

The exponent d_f characterizes the growth process and the resultant morphology of aggregate particles, and is regarded as a kind of fractal dimension.† Experimentally, the value of d_f can be determined by fitting the power law equation (5) to a set of data pairs (D_B, m) obtained for a number of aggregates at various stages of growth with a combined DMA-APM system. This method was proposed by K. Park of the University of Minnesota and his colleagues[35]: Using this method, they clarified how the fractal dimension of diesel exhaust particles varies as the engine load of diesel vehicles is increased. S. C. Kim, also of the University of Minnesota, and his colleagues showed that as the sintering temperature was increased from 20 ºC to 600 ºC, the fractal dimension of silver nano-particle aggregates varied from 2.07 to 2.95. [40] It is reasonable that the latter value is close to the fractal dimension of spherical particles of 3.

4.6 Dynamic shape factor[51][52]
The dynamic shape factor of a non-spherical particle, χ, is

defined as the ratio of its mobility B to the mobility B_ve of a spherical particle having the same volume as the particle under consideration. As the particle shape deviates more from the sphere, its dynamic shape factor, which is unity for a spherical particle, increases; hence it is used as a quantitative measure representing non-sphericity of the particle. The value of B_ve can be determined if its volume is inferred, for example, by the method mentioned in Subsection 4.3, while the mobility B of the particle can be conveniently measured with a DMA, and thus the value of χ can be derived from them.

J. Beranik of the Pacific Northwest National Laboratory, USA, and his colleagues presented a particle characterization method in which multiple particle properties including the dynamic shape factor are simultaneously measured with a combined system of an APM, a DMA, and a SPLAT (an instrument that analyzes chemical composition of individual aerosol particles).[51] L. Hillemann of the Technical University of Dresden and his colleagues experimentally investigated the change in the dynamic shape factor of particles produced at different sintering temperature.[52]

4.7 Porosity[53][54]
Porosity of a particle is defined as the ratio of the total volume of voids existent within or at the surface of the particle, V_v, to the volume surrounded by the envelope surface of the particle, V_e. If the envelope is approximately spherical, V_e can be derived from the mobility equivalent diameter. In addition, if the material of the particle is known, the particle volume excluding V_v can be derived from a measurement of the particle mass with an APM. The porosity can then be calculated as $V_\mathrm{v}/V_\mathrm{e}$. S. Y. Lee of Hiroshima University and his colleagues used this technique to investigate differences in the porosity of silica particles produced by several different methods.[53]

4.8 Specific surface area of individual particles[3][55]
It is known that if morphology and size of a particle satisfy certain loose conditions, the projected-area equivalent diameter of the particle can be approximated rather well by the mobility equivalent diameter.[56] This implies that the surface area of the particle should well correlate with the mobility equivalent diameter. A. D. Maynard, then at the Wilson Center, USA, and his colleagues proposed a new index of particle property based on this consideration, which is defined as

$$\Gamma = \pi D_B{}^2 / \tilde{m},\qquad(6)$$

where $\tilde{m}$ is the mode of the mass distribution of particles that have a given value of mobility equivalent diameter

---

† Because actual particles cannot have an exact self-similarity as defined mathematically, equation (5) holds true only approximately in limited ranges of m and D_B. The exponent d_f should rather be called mass-mobility exponent or fractal-like dimension. In the present article, however, we simply call it fractal dimension, for simplicity.

D_B.[3] Experimentally, the index Γ can be measured with a combined DMA and APM system.

Specific surface of powder represents the ratio of the total surface area to the total mass of a sample of powder as a whole, whereas the index Γ represents the ratio of the surface area to the mass of an individual particle having a given D_B. They demonstrated experimentally that the distribution of Γ and hence the physicochemical properties of single-walled carbon nanotubes (SWCNTs) vary significantly depending on the manufacturing process and production lot of the SWCNTs. They assert, on the basis of this observation, that such variations of particle properties should be taken into consideration when evaluating the potential hazards of nanoparticles.

5 From a concept to commercial products

Figure 5 shows an outline of the historical development leading to commercial products of the APM, divided crudely into three phases: the feasibility study phase, the problem solving phase, and the instrument development phase. The experimental part of the first phase research became possible only when it was funded by the Environment Agency of Japan (currently, the Ministry of the Environment) through a grant-in-aid for pollution protection that lasted from 1994 to 1998. When the prototype instrument constructed in this project was found to work roughly as we expected, we anticipated it would not be so difficult to develop an instrument with a sufficient practical performance. It turned out, however, that a couple of critical problems existed that hindered the instrument from working as we expected. Most of our efforts in the second phase research were devoted to solving these problems. For that, we received a grant from the New Energy and Industrial Technology Organization (NEDO), Japan, through the "Project on Basic Technologies for Nanomaterial Metrology" which lasted from 2001 to 2007. In what follows, development of each phase of the research is described from the standpoint of "synthesiology."

5.1 Feasibility study phase
The principle of the APM was devised by one of the authors

of this article, Ehara K., while he was a guest researcher at the Statistical Engineering Division (SED) of the National Institute of Standards and Technology (NIST), USA, from 1991 to 1992. While at NIST, he conducted statistically-designed experiments to evaluate the performance of a DMA. This work was supported by G. W. Mulholland at the Building and Fire Research Laboratory, NIST. A part of the findings obtained in this study was later published in a paper on the analysis of DMA data.[57]

In the course of this study, he became aware of the lack of a technique to measure mass of individual aerosol particles. Aerosol particles are in general non-spherical, and mobility of a non-spherical particle is a tensor depending on its orientation relative to the velocity. Unlike mobility, mass is an unambiguously defined physical property even for non-spherical particles, and is a strictly inherent property of the particle. The mass of aerosol particles should be worth measuring. The mass of atoms and molecules can be measured by the mass spectrometry, but if we try to apply its principle to aerosol particles, the instrument would become intolerably large in size, because the mass of typical aerosol particles is much larger than that of atoms and molecules. Further, a mass spectrometer operates only in a vacuum. If aerosol particles are brought into a vacuum, volatile materials in particles could produce change in their properties. Also, the concentration of particles when dispersed in a vacuum could become so low to detect the particles. Another principle which allowed us to measure mass of particles as they were suspended in the air was considered necessary. The principle described earlier in Subsection 3.2 was thus devised.

There were two good fortunes for Ehara, while he was staying at NIST. One was that the division he stayed was a group of scientists having expertise in mathematical statistics and probability theory. It is expected that when particles as small as 20 nm or below are to be measured, their Brownian motion can have non-negligible effects on measurement. He asked C. Hagwood and K. J. Coakley of the SED, NIST, to investigate its effects. A theoretical APM transfer function in
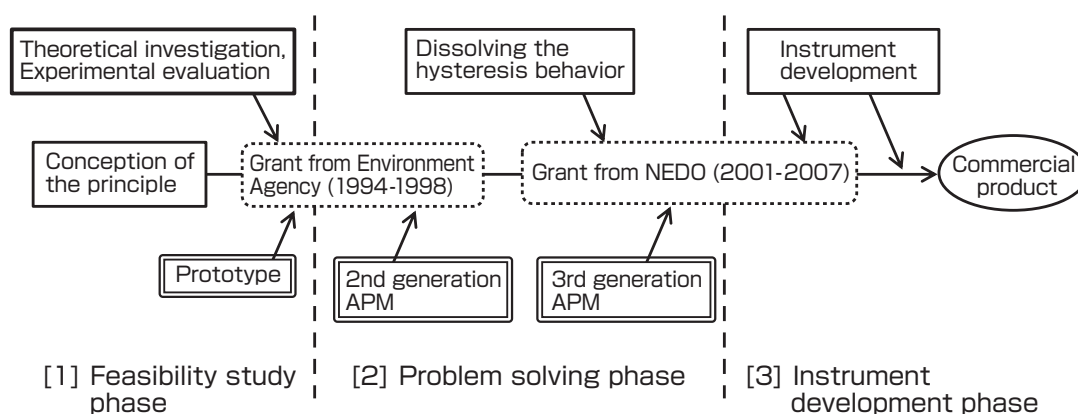

Fig. 5 Processes leading to commercial products of the APM.

which Brownian diffusion of the particles moving between the APM electrodes was taken into account was obtained in 1995, and this significant result led to the first paper regarding the APM.[58]

The other good fortune was that Fukushima N. of Kanomax Japan Inc., who was on a trip to the United States, had a visit to NIST. His visit was just by chance, but Ehara took this opportunity to consult with him about the feasibility of constructing an instrument based on the principle mentioned earlier. Fukushima had expertise in aerosol science as well as in engineering. He could grasp the significance of measurement of particle mass in aerosol science and technology, and also anticipate the difficulties in developing such an instrument.

Joint research to develop an APM started in 1994 between the National Research Laboratory of Metrology (NRLM; currently the National Metrology Institute of Japan, AIST) and Kanomax Japan, and a prototype instrument of APM was built in 1995. The mass distribution of monodisperse polystyrene latex (PSL) particles with a known diameter and density was measured, and a distinct peak in the APM spectrum corresponding to the PSL particles was successfully observed near the mass location expected theoretically.[6] This result convinced us of the feasibility of developing a practical instrument of the APM.

5.2 Problem solving phase

In designing the prototype, we recognized that it had some problems such as leakage of sample aerosols through the bearings. These problems were resolved in the improved second-generation instrument that was designed and built in 1996. The improved instrument had better resolution due to its higher maximum electrode rotational velocity. With this instrument, the peak locations in APM spectra observed for monodisperse PSL particles agreed well with those predicted from theory. However, in the course of extensive tests of its performance, two unexpected phenomena were

occasionally observed on the particle penetration rate. One was unexpectedly low reproducibility in particle penetration rates that could hardly be ascribed to statistical errors, and the other was peak heights in spectra significantly lower than those theoretically predicted. Both features were observed not consistently but on a rather unpredictable manner. Ehara and Fukushima tried for about a year to find the causes for such unexpected behavior of the APM, but did not succeed. In 1999, K. Worachotekamjorn of Prince of Songkla University, Thailand, stayed at AIST as a guest researcher for a year. During his stay, he took part in a more detailed experimental investigation on the unusual behavior of the APM. Also in 1999, Coakley of NIST had a short visit at AIST, and was engaged in a theoretical analysis of particle motion in the APM.

Experimentally, transient behavior of the particle penetration rate for monodisperse PSL particles was investigated. It revealed that the time required for the penetration rate to settle at a stationary value when the applied voltage was changed from a certain value to another was much longer than predicted theoretically. Strangely, the direction in which the penetration rate approaches a stationary value, i.e., whether the penetration rate increases or decreases during the transition time, was observed to be dependent on the previous value of the applied voltage. The experimental data indicated as "2nd generation" in Fig. 6(A) show transient behavior of the normalized penetration rate at the peak voltage of an APM spectrum, when the previous value of the voltage was lower than the peak voltage, while those in Fig. 6(B) show the transient behavior when the previous voltage was higher than the peak voltage.[59]

Theoretically, transient motion of particles within the APM electrodes when the applied voltage was changed stepwise was analyzed. Time-dependent stochastic differential equations were solved numerically, and the temporal variation of particle penetration rate was investigated. We were particularly concerned with the effect of slowly-moving particles present in the vicinity of the instrument walls,
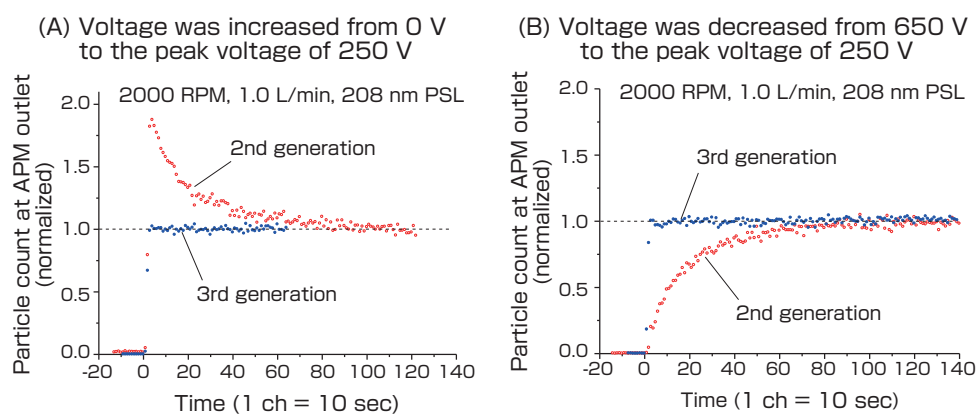

Fig. 6 Hysteresis observed in the temporal response of the second-generation APM, and that of the improved third generation.

as well as the effect of Brownian motion on the temporal variation of the particle penetration rate. Unfortunately, it turned out that neither of these effects explained the unexpected behavior of the penetration rate. The results obtained in this analysis, however, gave us clues narrowing down the causes for the unexpected phenomena, and helped us eventually reach the solution.

The attempt to find the cause continued for about two years in vain. However, in 2002, a plausible model that could explain the observed phenomena emerged. Particles that penetrate through the electrode gap carry charges of the same electrical polarity determined by the electrode configuration: When the outer electrode is set as the anode, the particles are positively charged, which we assume in the following scenario. The particles coming out of the electrodes enter a disk-shaped space leading to the outlet of the APM, which, in the second-generation APM, is surrounded by electrical insulators for isolation of the two electrodes. As shown in Fig. 7, when no voltage is applied, the particles pass through this space virtually without loss. When a certain voltage is applied, the particles experience an electrostatic field normal to the insulator surfaces, and thus some particles deposit on the surface of the insulator covering the cathode. As the number of deposited particles increases, Coulomb repulson by them increases accordingly and starts to push back the incoming particles. It takes a certain transient time for the surface density of the deposited particles and the number flux of the particles penetrating the space to converge to their equilibrium values. Note that these equilibrium values depend on the strength of

the applied voltage. This implies that when the applied voltage is varied abruptly from one value to another, the direction of convergence depends on the previous voltage setting. For example, if the voltage is set at V_{peak} (the peak voltage of an APM spectrum) from 0 V, the number of penetrating particles gradually decreases and converges to an equilibrium value, but when it is set at the same V_{peak} but from some higher voltage V_{high}, it gradually increases and reaches the same equilibrium value. In this way, this model could explain both the unexpectedly long transient time and the hysteresis behavior of the particle penetration rate as shown in Fig. 6.

On the basis of this model, the method of electrical insulation of the electrodes was modified so that particles experience no electrostatic field perpendicular to their flow direction outside the electrode gap. A third-generation APM with this modification was designed and constructed in 2003. We confirmed experimentally that the particle penetration rate behaved almost exactly as we theoretically expected, which is shown as the "3rd generation" data in Fig. 6.[60] We also found that the observed values of particle penetration rate agreed quite well with those theoretically predicted. These findings finally cleared the way toward an instrument having the expected performance. From the year 1997 when the unexpected behavior in particle penetration rate was found in the second-generation APM, it took us almost six years to eliminate these problems.

5.3 Instrument development phase[†]
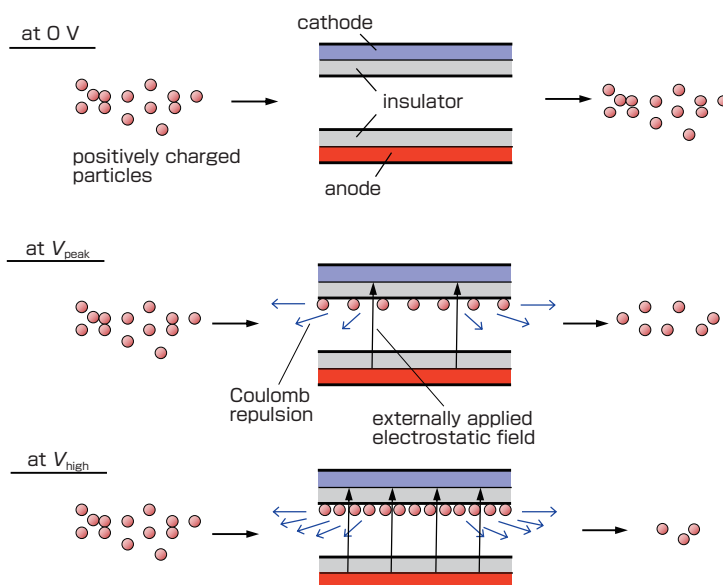After the unexpected behavior of the particle penetration

Fig. 7 A model of particle behavior in the disk-shaped space downstream of the electrode exit. Due to Coulomb repulsion from charged particles deposited on the insulator surface, the particle penetration rate through this space is lower than expected and dependent on the strength of the externally applied voltage.

† Certain commercial equipment, instruments, or materials are identified in this paper in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.

rate was cleared, we focused our efforts on development of instruments with improved accuracy, performance, and usability. Also, we conducted a study to apply the APM to characterization of diesel exhaust nanoparticles.[16][34] In the course of this study, Sakurai proposed the scanning mode operation of the APM, in which the applied voltage is varied not stepwise but continuously with time. Unlike the DMA for which the scanning mode operation is well-established, applicability of the scanning mode operation to the APM is not obvious, because particles cannot move straight through the electrode gap when the electrostatic field varies with time. Sakurai found that APM spectra obtained in the scanning mode operation under certain operating conditions can give almost the same measurement accuracy as the stepping mode operation.[61] This finding has enabled us to reduce the time to acquire one typical APM spectrum from approximately forty minutes down to five minutes.

The first commercial instrument, APM Model 3600, was developed by Kanomax Japan, and has been available since the year 2008. This instrument is capable of classifying particles in the mass range from 0.01 fg to 100 fg at a sample aerosol flow rate of 1 L/min. While conducting a detailed performance evaluation of this instrument, Tajima N. together with Sakurai devised the "APM operation diagram," which enables one to graphically select an operation condition optimum for a given particle mass.[7] Later, Tajima and her colleagues theoretically analyzed in detail how the design parameters of the APM affect its classification performance,[62] and on the basis of this analysis, a compact commercial instrument, APM Model 3601, was developed in 2012. The dimensions of the main body of this instrument, shown in Fig. 8, was 430 mm (width) by 200 mm (depth) by 140 mm (height). Though its standard flow rate of sample aerosols was set relatively small at 0.3 L/min, its classification performance was almost comparable to the Model 3600 APM. A newer model, APM Model 3602, with an improved control unit has replaced the APM Model 3601 and is commercially available at present.
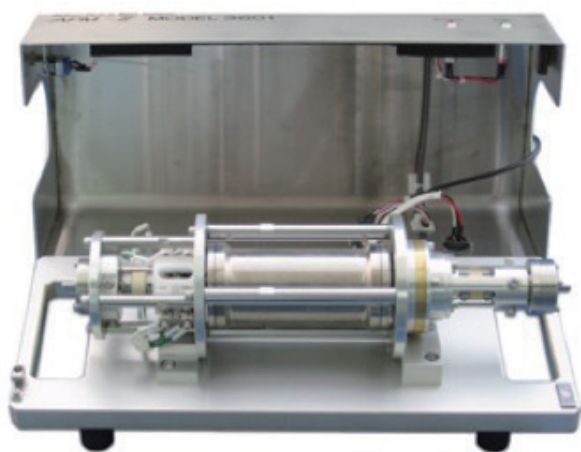

Fig. 8 Main part of the APM Model 3601.

5.4 Barrier to commercialization of an instrument

When some new technique is developed, its commercialization is essential for it to be utilized in society. However, to develop an apparently useful technique is one thing, and to render it a commercial product is another. It is usually difficult to anticipate to what extent a product will be accepted in the community relevant to the technique, especially when needs for the technique are not explicitly recognized in the community. A decision to commercialize the technique always poses some risk. There is a barrier between a seemingly useful technique and a commercial product based on it.

Just after the second-generation APM was built, P. H. McMurry of the University of Minnesota became interested in it, and introduced an instrument of the same make in his laboratory. Since then McMurry and his colleagues have published more than ten papers in which the APM was utilized in some ways. The University of Minnesota has been regarded as one of the centers of excellence in the community of aerosol science. The extensive use of the APM by McMurry and his colleagues has made the APM widely recognized in the community of aerosol science, which undoubtedly was a key factor to clear the barrier to a commercial product.

Ehara had a short visit to McMurry's laboratory in 1994, and several years later McMurry visited Ehara at the NRLM. Both visits had almost nothing to do with the APM, but Ehara had occasions to talk with McMurry about the plan and status of APM development. These unintended but fortunate occurrences played a critical role in the commercialization of the APM.

6 Conclusion

We have developed the aerosol particle mass analyzer (APM) that classifies aerosol particles according to their mass. In this article, we described, from the viewpoint of "synthesiology," the process of its development to its commercialization, as well as the work done by many other research groups to develop new techniques of aerosol particle characterization using the APM.

Expertise in aerosol instrumentation, expertise in applied mathematics to theoretically analyze the instrument performance, and generic knowledge of mechanical, electrical and fluid engineering to properly design an instrument, were all needed to develop a practically usable instrument. This naturally required involvement of many people having respective expertise. It was partly by luck that the participation of these people did occur during the process of developing the APM. Also, it was fortunate that we met a distinguished researcher who recognized the potential usefulness of the instrument while it was still under development.

There are still some issues that need to be addressed

regarding the APM. Installation of the scanning mode operation to the commercial models of the APM is yet to be completed. It might be possible to use the APM as the primary standard of particle mass in the metrological traceability system, because its working principle is the "zero method" which is usually expected to attain good measurement accuracy. Also, it is reported that below 30 nm, the diameter determined by the APM for spherical particles with a known density is consistently smaller than that determined by the DMA,[63] which needs to be investigated in more detail. In addition, a detailed performance evaluation of the CPMA, and realization of its theoretically expected performance are desired.

Acknowledgements

References

[1] P. Kulkarni, P. A. Baron and Klaus Willeke (eds.): *Aerosol Measurement: Principles*, Techniques, and Applications, 3rd edition, Wiley, Hoboken, N.J., (2011).

[2] K. Park, D. B. Kittelson, M. R. Zachariah and P. H. McMurry: Measurement of inherent material density of nanoparticle agglomerates, *J. Nanoparticle Res.*, 6 (2), 267–272 (2004).

[3] A. D. Maynard, B. K. Ku, M. Emery, M. Stolzenburg and P. H. McMurry: Measuring particle size-dependent physicochemical structure in airborne single walled carbon nanotube agglomerates, *J. Nanoparticle Res.*, 9 (1), 85–92 (2007).

[4] K. Ehara: Classification of airborne nanoparticles by size and mass, *Chemical Engineering*, 73 (2), 83–86 (2009) (in Japanese).

[5] E. O. Knutson and K. T. Whitby: Aerosol classification by electric mobility: apparatus, theory, and applications, *J. Aerosol Sci.*, 6 (6), 443–451 (1975).

[6] 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.*, 27 (2), 217–234 (1996).

[7] N. Tajima, N. Fukushima, K. Ehara and H. Sakurai: Mass range and optimized operation of the aerosol particle mass analyzer, *Aerosol Sci. Technol.*, 45 (2), 196–214 (2011).

[8] C. Presser, X. Ma, S. Guha, G. W. Mulholland and M. R. Zachariah: Online nanoparticle mass measurement by combined aerosol particle mass analyzer and differential mobility analyzer: Comparison of theory and measurements, *Aerosol Sci. Technol.*, 43 (11), 1075–1083 (2009).

[9] M. Kuwata: Particle classification by the tandem differential mobility analyzer-particle mass analyzer system, *Aerosol Sci. Technol.*, 49 (7), 508–520 (2015).

[10] G.-Y. Lin, B.-X. Liao, N.-J. Tzeng, C.-W. Chen, S.-N. Uang, S-C. Chen, D. Y. Pui and C. J. Tsai: The effect of nanoparticle convection-diffusion loss on the transfer function of an aerosol particle mass analyzer, *Aerosol Sci. Technol.*, 48 (6), 583–592 (2014).

[11] K. Park: In-situ measurements of physical properties of 50-500 NM particles: mass-mobility relationship, Ph. D. thesis, University of Minnesota, (2003).

[12] J. S. Olfert and N. Collings: New method for particle mass classification—the Couette centrifugal particle mass analyzer, *J. Aerosol Sci.*, 36 (11), 1338–1352 (2005).

[13] J. S. Olfert, K. StJ. Reavell, M. G. Rushton and N. Collings: The experimental transfer function of the Couette centrifugal particle mass analyzer, *J. Aerosol Sci.*, 37 (12), 1840–1852 (2006).

[14] E. Weingartner, H. Burtscher, C. Hüglin and K. Ehara: Semi-continuous mass measurement, in Aerosol Measurement: *Principles, Techniques, and Applications, Third Edition*, P. Kulkarni, P. A. Baron, K. Willeke eds., John Wiley & Sons, 255–268 (2011).

[15] 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.*, 36 (2), 227–238 (2002).

[16] K. Saito, O. Shinozaki, A. Yabe, T. Seto, H. Sakurai and K. Ehara: Measuring mass emissions of diesel particulate matter by the DMA-APM method (First Report): Measurement of the effective density of diesel exhaust particles, *Transactions of Society of Automotive Engineers of Japan*, 38 (6), 113–118 (2007) (in Japanese).

[17] Q. G. J. Malloy, S. Nakao, L. Qi, R. Austin, C. Stothers, H. Hagino and D. R. Cocker III: Real-time aerosol density determination utilizing a modified scanning mobility particle sizer—Aerosol particle mass analyzer system, *Aerosol Sci. Technol.*, 43 (7), 673–678 (2009).

[18] S. Nakao, M. Shrivastava, A. Nguyen, H. Jung and D. R. Cocker III: Interpretation of secondary organic aerosol formation from diesel exhaust photooxidation in an environmental chamber, *Aerosol Sci. Technol.*, 45 (8), 964–972 (2011).

[19] T. L. Barone, A. A. Lall, J. M. E. Storey, G. W. Mulholland, V. Y. Prikhodko, J. H. Frankland, J. E. Parks and M. R. Zachariah: Size-resolved density measurements of particle emissions from an advanced combustion diesel engine: Effect of aggregate morphology, *Energy Fuels*, 25 (5), 1978–1988 (2011).

[20] X. Tang, D. R. Cocker III and A. Asa-Awuku: Are sesquiterpenes a good source of secondary organic cloud condensation nuclei (CCN)? Revisiting β-caryophyllene CCN, *Atmospheric Chem. Phys.*, 12 (18), 8377–8388 (2012).

[21] S. H. Kim, G. W. Mulholland and M. R. Zachariah: Density measurement of size selected multiwalled carbon nanotubes by mobility-mass characterization, *Carbon*, 47 (5), 1297–1302 (2009).

[22] M. Kuwata, S. R. Zorn and S. T. Martin: Using elemental ratios to predict the density of organic material composed of carbon, hydrogen, and oxygen, *Environ. Sci. Technol.*, 46 (2), 787–794 (2012).

[23] H. Sakurai, K. Park, P. H. McMurry, D. D. Zarling, D

B. Kittelson and P. J. Ziemann: Size-dependent mixing characteristics of volatile and nonvolatile components in diesel exhaust aerosols, *Environ. Sci. Technol.*, 37 (24), 5487–5495 (2003).

[24] N. Moteki and Y. Kondo: Effects of mixing state on black carbon measurements by laser-induced incandescence, *Aerosol Sci. Technol.*, 41 (4), 398–417 (2007).

[25] A. A. Lall, W. Rong, L. Mädler and S. K. Friedlander: Nanoparticle aggregate volume determination by electrical mobility analysis: Test of idealized aggregate theory using aerosol particle mass analyzer measurements, *J. Aerosol Sci.* 39 (5), 403–417 (2008).

[26] M. Kuwata and Y. Kondo: Measurements of particle masses of inorganic salt particles for calibration of cloud condensation nuclei counters, *Atmospheric Chem. Phys.*, 9 (16), 5921–5932 (2009).

[27] M. Kuwata, Y. Kondo and N. Takegawa: Critical condensed mass for activation of black carbon as cloud condensation nuclei in Tokyo, *J. Geophys. Res. Atmospheres*, 114 (D20) (2009).

[28] M. Shiraiwa, Y. Kondo, T. Iwamoto and K. Kita: Amplification of light absorption of black carbon by organic coating, *Aerosol Sci. Technol.*, 44 (1), 46–54 (2010).

[29] X. Ma, A. A. Lall, G. W. Mulholland and M. R. Zachariah: Evaporation anisotropy of free nanocrystals, *J. Phys. Chem. C*, 115 (34), 16941–16946 (2011).

[30] S. Guha, X. Ma, M. J. Tarlov and M. R. Zachariah: Quantifying ligand adsorption to nanoparticles using tandem differential mobility mass analysis, *Anal. Chem.*, 84 (15), 6308–6311 (2012).

[31] M. Laborde, P. Mertes, P. Zieger, J. Dommen, U. Baltensperger and M. Gysel: Sensitivity of the single particle soot photometer to different black carbon types, *Atmospheric Meas. Tech.*, 5 (5), 1031–1043 (2012).

[32] J. G. Radney, X. Ma, K. A. Gillis, M. R. Zachariah, J. T. Hodges and C. D. Zangmeister: Direct measurements of mass-specific optical cross sections of single-component aerosol mixtures, *Anal. Chem.*, 85 (17), 8319–8325 (2013).

[33] K. Park, D. B. Kittelson and P. H. McMurry: A closure study of aerosol mass concentration measurements: Comparison of values obtained with filters and by direct measurements of mass distributions, *Atmos. Environ.*, 37 (9), 1223–1230 (2003).

[34] K. Saito, O. Shinozaki, A. Yabe, T. Seto, H. Sakurai and K. Ehara: Measuring mass emissions of diesel particulate matter by the DMA-APM method (Second Report): Comparison with the gravimetric filter method, Transactions of Society of Automotive Engineers of Japan, 39 (4), 97–102 (2008) (in Japanese).

[35] K. Park, F. Cao, D. B. Kittelson and P. H. McMurry: Relationship between particle mass and mobility for diesel exhaust particles, *Environ. Sci. Technol.*, 37 (3), 577–583 (2003).

[36] B. K. Ku, M. S. Emery, A. D. Maynard, M. R. Stolzenburg and P. H. McMurry: In situ structure characterization of airborne carbon nanofibres by a tandem mobility-mass analysis, *Nanotechnology*, 17, 3613–3621 (2006).

[37] J. S. Olfert, J. P. R. Symonds and N. Collings: The effective density and fractal dimension of particles emitted from a light-duty diesel vehicle with a diesel oxidation catalyst, *J. Aerosol Sci.*, 38, 69–82 (2007).

[38] R. Zhang, A. F. Khalizov, J. Pagels, D. Zhang, H. Xue and P. H. McMurry: Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing, *Proc. Natl. Acad. Sci.*, 105, 10291–10296 (2008).

[39] W. G. Shin, G. W. Mulholland, S. C. Kim, J. Wang, M. S. Emery and D. Y. H. Pui: Friction coefficient and mass of silver agglomerates in the transition regime, *J. Aerosol Sci.*, 40 (7), 573–587 (2009).

[40] S. C. Kim, J. Wang, M. S. Emery, W. G. Shin, G. W. Mulholland and D. Y. H. Pui: Structural property effect of nanoparticle agglomerates on particle penetration through fibrous filter, *Aerosol Sci. Technol.*, 43 (4), 344–355 (2009).

[41] J. Pagels, A. F. Khalizov, P. H. McMurry and R. Y. Zhang: Processing of soot by controlled sulphuric acid and water condensation—Mass and mobility relationship, *Aerosol Sci. Technol.*, 43 (7), 629–640 (2009).

[42] N. Moteki, Y. Kondo, N. Takegawa and S. Nakamura: Directional dependence of thermal emission from nonspherical carbon particles, *J. Aerosol Sci.*, 40 (9), 790–801 (2009).

[43] J. H. Scheckman, P. H. McMurry and S. E. Pratsinis: Rapid characterization of agglomerate aerosols by in situ mass-mobility measurements, *Langmuir*, 25 (14), 8248–8254 (2009).

[44] W. G. Shin, G. W. Mulholland and D. Y. H. Pui: Determination of volume, scaling exponents, and particle alignment of nanoparticle agglomerates using tandem differential mobility analyzers, *J. Aerosol Sci.*, 41 (7), 665–681 (2010).

[45] J. H. Scheckman and P. H. McMurry: Deposition of silica agglomerates in a cast of human lung airways: Enhancement relative to spheres of equal mobility and aerodynamic diameter, *J. Aerosol Sci.*, 42 (8), 508–516 (2011).

[46] T. Torvela, A. Lähde, J. Mönkäre, J. Riikonen and J. Joutsensaari: Low-temperature aerosol flow reactor method for preparation of surface stabilized pharmaceutical nanocarriers, *J. Aerosol Sci.*, 42 (10), 645–656 (2011).

[47] M. L. Eggersdorfer, D. Kadau, H. J. Herrmann and S. E. Pratsinis: Aggregate morphology evolution by sintering: Number and diameter of primary particles, *J. Aerosol Sci.*, 46, 7–19 (2012).

[48] M. Shapiro, P. Vainshtein, D. Dutcher, M. Emery, M. R. Stolzenburg, D. B. Kittelson and P. H. McMurry: Characterization of agglomerates by simultaneous measurement of mobility, vacuum aerodynamic diameter and mass, *J. Aerosol Sci.*, 44, 24–45 (2012).

[49] J. Olfert and S. Rogak: Universal relations between soot effective density and primary particle size for common combustion sources, *Aerosol Sci. Technol.*, 53 (5), 485–492 (2019).

[50] A. Schmidt-Ott, U. Baltensperger, H. W. Gäggeler and D. T. Jost: Scaling behaviour of physical parameters describing agglomerates, *J. Aerosol Sci.*, 21 (6), 711–717 (1990).

[51] J. Beranek, D. Imre and A. Zelenyuk: Real-time shape-based particle separation and detailed in situ particle shape characterization, *Anal. Chem.*, 84 (3), 1459–1465 (2012).

[52] L. Hillemann, F. Babick and M. Stintz: Measurement of the dynamic shape factor using APM and SMPS in parallel, *Procedia Eng.*, 102, 1177–1182 (2015).

[53] S. Y. Lee, H. Chang, T. Ogi, T. Iskandar and K. Okuyama: Measuring the effective density, porosity, and refractive index of carbonaceous particles by tandem aerosol techniques, *Carbon*, 49 (7), 2163–2172 (2011).

[54] Q. Liu, X. Ma and M. R. Zachariah: Combined on-line differential mobility and particle mass analysis for determination of size resolved particle density and microstructure evolution, *Microporous Mesoporous Mater.*, 153, 210–216 (2012).

[55] B. K. Ku and D. E. Evans: Investigation of aerosol surface

area estimation from number and mass concentration measurements: Particle density effect, *Aerosol Sci. Technol.*, 46 (4), 473–484 (2012).

[56] S. N. Rogak, R. C. Flagan and H. V. Nguyen: The Mobility and structure of aerosol agglomerates, *Aerosol Sci. Technol.*, 18 (1), 25–47 (1993).

[57] K. Ehara, G. W. Mulholland and R. C. Hagwood: Determination of arbitrary moments of aerosol size distributions from measurements with a differential mobility analyzer, *Aerosol Sci. Technol.*, 32 (5), 434–452 (2000).

[58] C. Hagwood, K. Coakley, A. Negiz and K. Ehara: Stochastic modeling of a new spectrometer, *Aerosol Sci. Technol.*, 23 (4), 611–627 (1995).

[59] K. Worachotekamjorn and K. Ehara: Performance evaluation of the aerosol particle mass analyzer, *Proc 1st Asian Part. Technol. Symp. APT 2000 Bangk.*, S-VI-1 (2000).

[60] K. Ehara, K. Fukushima, K. Worachotekamjorn and K. J. Coakley: Analysis and improvement of the temporal response of the aerosol particle mass analyzer, *J. Aerosol Sci.*, 35, Supplement 1 (Abstracts of European Aerosol Conference) (2004).

[61] H. Sakurai and K. Ehara: Ryushi shitsuryo bunseki hoho (Particle mass analysis method), Unexamined Patent Application 2008-224246 (2008) (in Japanese).

[62] N. Tajima, H. Sakurai, N. Fukushima and K. Ehara: Design considerations and performance evaluation of a compact aerosol particle mass analyzer, *Aerosol Sci. Technol.*, 47 (10), 1152–1162 (2013).

[63] H. Sakurai, K. Ehara, N. Tajima and N. Fukushima: Investigation on the cause for underestimation of density measured by the DMA-APM method, *Poster Presentation at 2010 Annual Conf. Am. Assoc. Aerosol Res., Portland Or.* (2010).

Authors

Ehara Kensei

Ehara Kensei received a B.S. in Physics from Kyoto University in 1978, and a Ph.D. in Physics from Osaka University in 1983. In the same year, he joined the National Research Laboratory of Metrology, which currently is the National Metrology Institute of Japan (NMIJ) of the National Institute of Advanced Industrial Science and Technology (AIST). There he had been engaged in application of statistics to metrology, development of particle mass and size standards, and research on aerosol instrumentation. He retired from AIST in 2019. Currently, he is Researcher Emeritus of AIST, and teaches at Meiji University and Gakushuin University as an adjunct professor. He has been engaged in development of the APM from its beginning.

Charles Hagwood

Charles Hagwood received a B.S. degree in mathematics from NC A&T State University, Greensboro, NC, USA, and a Ph.D. degree in mathematics from the University of Michigan, Ann Arbor, MI, USA. He taught in the Department of Mathematics, Dartmouth College, Hanover, NH, USA, and with the Department of Mathematics, University of Virginia, Charlottesville, VA, USA. He is with the National Institute of Standards and Technology (NIST). As a Ford Foundation Fellow, he spent one year with the Department of Statistics, Stanford University, Stanford, CA, USA. He is currently with the Statistical Engineering Division, NIST. His current research and consulting involve stochastic differential equations applied to sizing aerosol particles, shape analysis, dynamic spectrum access, standard reference materials and analytical chemistry, and point process modeling. In the research presented in this article, he theoretically analyzed the effect of Brownian motion of particles on the APM transfer function. He is the lead author of the first paper that was ever written about the APM.

Kevin J. Coakley

Kevin J. Coakley earned a BS in Physics from Yale University, an MS in Physics from the University of Washington (Seattle) and a PhD in Statistics from Stanford University. He joined the Statistical Engineering Division at the National Institute of Standards and Technology (NIST) in 1989. Current research interests include stochastic modeling, planning and analysis of experiments in physics, statistical methods for broadband microwave imaging of materials and atom probe tomography, and statistical learning methods for physical science applications including Johnson Noise Thermometry. He was engaged in theoretical analysis of the effects of Brownian motion and transient behavior of particles on the APM performance.

Fukushima Nobuhiko

Fukushima Nobuhiko graduated from the Department of Chemical Engineering, Graduate School of Engineering, Kanazawa University in 1978. He joined Nihon Kagaku Kogyo K.K. (currently, Kanomax Japan Inc.) in 1978. He had been engaged in R&D for fine particle measurement. He received a Ph.D. in Chemical Engineering from the University of Osaka Prefecture in 1995. In Kanomax Japan, he served as Director in charge of Production from 1998, Vice President from 2007, Senior Executive Officer and Vice President from 2008, and is currently Executive Vice Chairman. He has been engaged in development of the APM from its initial stage, and engineered the prototype and commercial instruments of the APM.

Kittichote Worachotekamjorn

Kittichote Worachotekamjorn graduated in 1992 from the Department of Pharmaceutical Technology, Faculty of Pharmaceutical Sciences, Prince of Songkla University (PSU), Southern Thailand. He joined Faculty of Pharmaceutical Science, PSU, as Lecturer at the Department of Pharmaceutical Technology since 1992. He got a JICA scholarship, and stayed at the National Research Laboratory of Metrology in Tsukuba, Japan, as a guest researcher during 1999–2000. He was engaged in experimental evaluation of the performance of the APM, and clarified the details of the

transient behavior of the particle penetration rate.

Tajima Naoko

Tajima Naoko graduated from the Department of Chemical Engineering, Graduate School of Engineering, Kanazawa University in 1984. She joined Nippon Kagaku Kogyo K.K. (currently, Kanomax Japan Inc.) in 1984. While working there, she completed a Ph.D. program and received a Ph.D. degree in Chemical Engineering from the Department of Chemical Engineering, Graduate School of Engineering, Hiroshima University in 2013. She joined the Nanoelectronics Research Institute in the National Institute of Advanced Industrial Science and Technology (AIST) in 2014, and is currently Project Researcher at the CNT-Application Research Center, AIST. She was engaged in development and performance evaluation especially of commercial models of the APM. She conducted theoretical and experimental analyses of the APM performance that led to the compact model of the APM.

Sakurai Hiromu

Sakurai Hiromu received his Ph.D. degree in Chemistry from the Pennsylvania State University in 1999. He conducted research at the University of Minnesota in 1999–2003, and at the National Institute of Advanced Industrial Science and Technology (AIST) in 2003, both as a post-doctoral researcher. He joined AIST in 2004, and had led the Particle Measurement Section of the National Metrology Institute of Japan (NMIJ), AIST, since 2013. He is currently the Leader of the Particle Measurement Group, NMIJ, AIST. In the research presented in this article, he proposed the scanning mode operation, as well as a graphical tool to select an optimal operating condition of the APM. He also developed methods of applying the APM for characterization of airborne nanoparticles including diesel exhaust particles and carbon nanotubes.

Discussions with Reviewers

1 Overall
Comment (Fujii Kenichi, AIST)

There are rising interests in health, environment, and safety, and the importance of evaluation of fine particles is widely recognized in the industrial field such as in clean room management. Amidst this background, I think this is a high quality paper in the points that it describes the measurement principle of the aerosol particle mass analyzer (APM) that the authors created to measure aerosol particles in micromass range that could not be measured by conventional methods, and it presents case studies in which the developed device was used for evaluation of various particle properties.

It is extremely innovative that this method enables evaluation of aerosol particle mass in the range from 3×10^{-18} to 2×10^{-12} g that could not be covered by conventional measurement technologies, including minimum 0.1 µg for mass measurement by an electronic balance, 10^{-11} g to 10^{-5} g that is the mass measurement range of a tapered element oscillating microbalance (TEOM), and 1.7×10^{-24} g to 1.7×10^{-18} g that is the mass measurement range by time-of-flight mass spectrometry (TOF-MS).

The definition of the kilogram was revised on May 20, 2019 for the first time in 130 years, and the shift was made from a definition by an artifact to a definition by Planck constant. In the field of mass standard, there is a growing demand for the measurement in the range of 0.1 µg or less that could not be measured before. I think it is a timely paper on pioneering a new measurement range.

Comment (Ichimura Shingo, Waseda University)

This paper is a compact summary from the principle to device development of the aerosol particle mass analyzer (APM), overcoming of various issues in practical application and product development, and development of various particle property evaluation technologies (mainly by others) as ripple effects of APM. I think it is a paper appropriate for publication in *Synthesiology* whose objective is "to describe the goal of research and its social value, the scenario and research procedure for achieving the goal, and the process of integrating the elemental technologies."

2 Measurement principle of APM and its uncertainty
Question 1 (Fujii Kenichi)

In Subsection 3.2, you explain the principle of mass measurement of aerosol particles using APM. I understand that the particles are classified by the ratio of mass and charge of the charged particles (mass-to-charge ratio) obtained by applying voltage between the inner and outer cylinders that rotate at the same angle speed. However, there is no description about measurement parameters or derived equations needed to specifically calculate particle mass. I think the readers will be able to understand this measurement principle more deeply if you explain specifically.

Answer 1 (Ehara Kensei)

The classification performance of the APM is characterized by the APM transfer function, and an experimental spectrum obtained by the APM can be theoretically represented by an integral involving the transfer function. I have added a brief description of the transfer function in Subsection 3.2. For more details about the transfer function, please refer to the references cited.

Question 2 (Fujii Kenichi)

You explain that mass measurement by APM is not the measurement of mass of a single particle, but is measurement of mass distribution. Can you also provide some representative example? It can be an example in which you obtained the highest precision. Can you also mention your thoughts on uncertainty in the measurement of mass distribution, relative uncertainty, and causes of uncertainty?

Answer 2 (Ehara Kensei)

The electro-gravitational aerosol balance (EAB) is a method that we have developed for absolute measurement of particle mass which operates in much the same way as the APM [K. Ehara, K. Takahata, and M. Koike, *Aerosol Sci. Technol.* 40, 514–520 (2006) and *Aerosol Sci. Technol.* 40, 521–535 (2006); K. Takahata, H. Sakurai, and K. Ehara, *Aerosol Sci. Technol.* 54, *-* (2020)]. The EAB uses gravitational force instead of centrifugal force, and provides measurement accuracy better than the APM, though it lacks practicality in measurement due to its significantly long measurement time required. Because a detailed uncertainty analysis has been conducted for the EAB, we present it first: The relative expanded uncertainty with a coverage factor of two of the number average diameter of 100 nm PSL particles (approximately 0.57 fg in mass) is 0.66 % (or 1.9 % in terms of the number average mass). We have confirmed that particle mass obtained with the APM is within ±5 % of that obtained with the EAB (see Reference [7]).

Comment 3 (Fujii Kenichi)

In Subsection 3.3, there is an explanation of the Couette centrifugal particle mass analyzer (CPMA) in which the inner electrode is designed to rotate faster than the outer electrode. You use the terminology "Couette flow." I think you should provide an explanation in the footnote to enable the readers to understand the

characteristic of this measurement principle.

Answer 3 (Eʜᴀʀᴀ Kensei)

We have added an explanation of the Couette flow as a footnote in Subsection 3.3.

3 Reliability of mass measurement by TEOM

Question 1 (Fᴜᴊɪɪ Kenichi)

You describe in Subsection 3.4 that mass measurement using a tapered element oscillating microbalance (TEOM) covers the range from 10^{-11} g to 10^{-5} g. I think the reason it is difficult to maintain traceability with TEOM is because it utilizes the oscillation principle, which makes it difficult to calibrate the relation between mass and oscillation. Can you please briefly explain, how TEOM normally maintains its traceability to the mass standard?

Answer 1 (Sᴀᴋᴜʀᴀɪ Hiromu and Eʜᴀʀᴀ Kensei)

The TEOM has a disposable filter cartridge for collection of particles attached to the tip of the oscillating element. A sample aerosol is continuously drawn through the filter, and the mass of the particles collected on the filter is obtained from the change in the characteristic frequency of the oscillator. Calibration is done by measuring the characteristic frequencies when a filter cartridge with a precisely known mass is attached to and detached from the oscillating element.

Comment and question 2 (Fᴜᴊɪɪ Kenichi)

In the measurement of particle mass by TEOM, the relationship of mass and natural frequency is obtained from the change of natural frequency when the filter, whose mass has been calibrated using a balance, is installed and removed. This is described in a number of documents. However, in this case, the minimum value (0.1 μg) of mass measurement by a balance will end up extrapolated to a smaller range by the oscillation principle, and I imagine that the degree to which this extrapolation is correct will become a problem. Since the particles are trapped in the filter attached to the oscillator, if there is some kind of mechanical coupling linked by spring in the system of the oscillator, filter, and particle, there may be a possibility that the mass of particles will not be correctly reflected in the natural frequency. Therefore, there is a question of whether correct extrapolation can be done to 10 pg, and in the field of mass standard, there are not infrequent doubts expressed on the reliability of mass measurement by the principle of oscillation.

On the other hand, mass measurement based on the principle of oscillation has extremely high sensitivity, and it is a fact that this principle is being actively applied in some fields, particularly in the field of MEMS.

You show the range of mass measurement by TEOM in Fig. 4, and I think the reliability will increase if one can compare the measurement results of TEOM and APM per 1 pg. Are there past cases that investigated whether the results measured matched those by methods with different principles?

Answer 2 (Eʜᴀʀᴀ Kensei)

There is a paper in which a particle diameter determined from the mass measured with the EAB (see Answer 2 in Discussion 2 for the EAB) for particles with a known density was compared with diameters measured by methods based on other measurement principles [T. A. Germer *et al.*, *Proc. SPIE.*, 4779:60–71 (2002)]. It reports that the diameter of 100 nm PSL particles we obtained with the EAB (100.8 nm ± 0.67 nm) and that of the same particles obtained by NIST using a DMA with rigorous metrological traceability agreed quite well. Good agreement between the APM and the EAB was already mentioned in Answer 2 of Discussion 2. We believe that these results indicate that the EAB as well as the APM has measurement accuracy of the level that we expect.

It might be possible to examine the reliability of the TEOM by generating monodispersed particles, the mass of which is determined beforehand by the EAB or APM, in the atmosphere at a known concentration, and sending them to the TEOM. We

are, however, not familiar with the TEOM, and do not know for sure whether such a method works as expected. Also, we were unfortunately unable to get information on how the reliability of the TEOM is ensured.

4 Treatment of intellectual property in research aiming at commercialization of a product

Question and comment 1 (Iᴄʜɪᴍᴜʀᴀ Shingo)

When aiming for product realization starting from the principle of a new measurement device, in general, one of the main targets is the creation of intellectual property (such as a patent). In this paper, there is no description from that perspective.

I think it will develop readers' thinking if you add how you handled intellectual property in conducting this research (if you purposefully did not consider acquiring intellectual property, can you explain why). Please consider adding something about this.

Answer 1 (Eʜᴀʀᴀ Kensei)

I did not refer to the intellectual property. I have added a text regarding the patent application in Subsection 5.1.

Question and comment 2 (Iᴄʜɪᴍᴜʀᴀ Shingo)

I read your answer to Comment 1. As you indicated, you added a text about the creation of intellectual property in the conceptual phase (description about basic patent application in Japan and US). However, the main point of my comment was to clarify what kind of thought process there was as a public research institution, in conducting joint research with a private company to improve the device, and thereby achieving the final practical application (product realization). For example, in the following texts, there are several indications of the potential for creating intellectual property toward product realization:

> Page 11
> "the first prototype of APM was completed in 1995."
> Page 12
> "the third-generation device in which revisions were made so the problematic phenomena did not occur by devising the insulation of positive and negative electrodes was created in 2003."
> Page 14
> "the scanning mode operation in which the electrode voltage was continuously changed as the function of time rather than in steps"
> Page 14
> "'APM operation diagram,' a tool for selecting the optimal operation condition according to the particle mass"

In this case, I think it is beneficial to the reader who are engaging in *Full Research*, if you further describe what thoughts you had for the obtainment of intellectual property (in some cases, secret know-how) toward product realization, in conducting joint research. Please consider.

Answer 2 (Eʜᴀʀᴀ Kensei)

I agree that treatment of intellectual property is an important subject in "synthesiology," and think that a suitable strategy is needed to handle it. However, it is hard for us to say that our research has been pursued with sufficient recognition of the importance of the intellectual property. We do not have anything to say about it in our article.

For now, I think that patent application by a public institution like us should be as restrictive as possible, for example by limiting it to a principle patent, so that the intellectual property be shared widely in society. The patent application for the APM proceeded effectively in that way. In the case of the DMA (see Subsection 3.2), on the other hand, an instrument with a design very close to the currently prevalent model was developed in the 1950s, but a patent was not filed for it. I think the lack of a patent

made the DMA easier for many researchers to choose it as a research subject, and as a result, various improvements of the instrument had advanced. It might have been a reasonable idea not to file a patent for the APM, but without a patent protection it could be difficult for a private company to proceed to its commercialization. The intellectual property must be handled carefully with due consideration to the relevant market size and various risks associated with its commercialization. Looking back the process of APM development, I still do not know what was the right decision with regard to handling the intellectual property. I would welcome any of your advice on it.

Additional question and comment (Iᴄʜɪᴍᴜʀᴀ Shingo)

I read your answer to my question. I think by making your statement in the text, you will give this paper synthesiological consideration. For example, I think one of the proposals is to add Subsection 5.5 "Thinking about the intellectual property in R&D," but since this will affect the structure of the paper, I shall leave the decision to the authors.

Answer (Eʜᴀʀᴀ Kensei)

Please allow us to leave the discussion on intellectual property above in this "Discussion with Reviewers" section.

5 Future development

Comment 1 (Fᴜᴊɪɪ Kenichi)

On May 20, 2019, the definition of the kilogram shifted to the Planck constant. The traceability of mass measurement can be achieved by tracing mass *m* of the object to Planck constant *h*. Therefore, in the "mise en pratique" for the definition of the kilogram prepared by the Consultative Committee for Mass and Related Quantities (CCM) of the International Committee of Weights and Measures (CIPM), the watt (Kibble) balance method and the X-ray crystal density method are described as representative measurement methods. In principle, the "mise en pratique" states that as long as the ratio *h*/*m* is measured with traceability, any method can be used. In the case of APM, what is the route of traceability to *h*? A conceptual explanation will be fine. What must be made traceable?

Also, I think there is the possibility of measuring micromass using elementary electric charge *e* or Boltzmann constant *k* that were newly defined as SI. In the case of aerosol particles, for example, please mention any elemental technology that may be useful in the future, as well as the future direction of the development of measurement principles.

Answer 1 (Eʜᴀʀᴀ Kensei)

First, let me describe the traceability of the EAB (see Answer 2 in Discussion 2), because its measurement principle is significantly simpler than that of the APM. Let us assume, for simplicity, that the sample particles have a uniform mass *m* and are singly-charged. Then the EAB can be regarded as measurement of the voltage *V* applied between plate electrodes that realizes balance between electrostatic force *eV*/*H* (*e* is the elementary charge, and *H* is the electrode gap) and gravity *mg* (g is the gravitational acceleration) experienced by the particles. From the equation representing the force balance, the particle mass is given by

$$m = \frac{eV}{gH}. \tag{A}$$

To be strict, the mass distribution of real particles has a finite width, and accordingly this equation as it stands is not used to determine the number average mass in the EAB method, but is still valid as far as the measurement traceability is concerned. In the current SI, which has undergone the 2019 revision, *e* is a defined value; *V* is traceable to *e*, *h* (the Planck constant), and Δv_{Cs} (the transition frequency of the caesium 133 atom) via the Josephson voltage standard; and *H* and g are both traceable to c (the speed of light) and Δv_{Cs}. Accordingly, the particle mass m is ultimately traceable to the four constants, *e*, *h*, *c*, and Δv_{Cs}.

In the case of the APM, *g* in equation (A) is replaced with the centrifugal acceleration associated with the electrode rotation, and the strength of the electrostatic field *V*/*H* with that relevant to the cylindrical electrodes, but the traceability paths are the same.

I think we should not simply expect that the recent revision of the SI, particularly that of the definition of the kilogram, can naturally bring some benefit to measurement of mass of tiny objects such as particles. One of the reasons for this is that the mass of particles obtained with the APM or EAB already is not directly traceable to the kilogram prototype in the previous version of the SI, but is traceable to the Planck constant *h* and elementary charge *e* via the voltage standard, as explained above. The fact that these constants have zero uncertainties in the revised SI may, in principle, lead to a reduction of uncertainty in particle mass measurement, but the uncertainty components associated with these constants were already negligible in the framework of the former SI. This means that the fact the kilogram was defined as the mass of a somewhat unstable macroscopic artifact, i. e., the international kilogram prototype, did not pose any practical obstacles already in the framework of the previous version of the SI.

The recent revision of the SI has brought some obvious benefits to measurement of microscopic entities such as elementary particles. However, all such benefits are in the reduction of measurement uncertainties, and we cannot naively expect that the revision in the definition of the kilogram will bring us a new measurement technique which can cover a mass range that has not been covered by the existing techniques. I hope a new superior technique of mass measurement which relies essentially on the revised definition of the kilogram will emerge, but I think it will take a long time, possibly a very long time, for this to happen.

Comment 2 (Fᴜᴊɪɪ Kenichi)

Thank you for presenting your thoughts on traceability of particle mass measurement by EAB and APM. In the SI definition revision including the kilogram, since the uncertainty of Planck constant *h* became zero, the uncertainty of the fine structural constant as well as the mass of atom and elementary particles certainly decreased by applying the principle of atomic interference. In the revision of SI, it is considered important to provide a definition that is applicable to future technological innovations, not just to reduce current uncertainties. As a result of defining the length by speed of light, several technological innovations were born in the field of optical frequency measurement. I hope there will be further technological innovations in the field of particles and aerosols, triggered by the development of the APM.