Simple Method of Size Estimation of Radioactive Aerosols in Air Monitoring

Yuji Yamada*1 and Akira Koizumi**1

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A new simplified method for particle size estimation of radioactive aerosols was proposed for air monitoring. It does not need any new instruments but a traditional aerosol sampling device with a multistage filter system. The method is based on the particle size dependency of penetration for air filter. Decontamination factor data measured in each filter stage is inversed to a size distribution of aerosols. When air filter for a dust sampler such as Toyo HE-40T is used, the method can be applied for a size estimation of aerosols ranging from 0.2 to 10 μm in which ICRP is interested for lung deposition model. In a size measurement of 198Au aerosols, it was shown that the size distribution estimated by the present method was in a good agreement with the distribution by the existent method using a cascade impactor.

KEY WORDS: particle size estimation, radioactive aerosol, air monitoring, multistage filtration, decontamination factor

I INTRODUCTION

In an accidental case of inhalation of radioactive aerosols, information on the particle size as well as the inhaled radioactivity, radionuclide and chemical form is of very importance for assessment of its biological effects.

Most of dust samplers used in nuclear facilities is equipped with a filter system, which makes it easy for health physicists to get the above informations to some extent. However, it is not easy to know the particle size from the filter sample, though cascade impactor type samplers such as an Andersen sampler serve the purpose entirely. Other than this, Leary determined particle size by the autoradiography,1) and recently Iida et al. modified it to speedy method by using a micro-channel plate image intensifier.2) But these methods need complicated techniques and special instruments for analysis, and samples are limited to aerosols with a high specific activity. Therefore, it is not always appropriate for health physicists to use them in air monitoring. We propose a new simplified method, multistage filtration (MSF) method, which is based on a particle size dependency of its penetration through a filter.

In this paper, we report on the MSF method and discuss the practicability to field measurement, giving an example of comparative study for validation of the method.

II MULTISTAGE FILTRATION THEORY

Particle penetration through a fibrous filter depends on a particle size. The particle size distribution in downstream of the filter is not the same as in upstream, which means that in multistage filter system, the size distribution in upstream of filter is different in every filter stage. Consequently, DF (Decontamination Factor; defined as a ratio of radioactivity in upstream of
filter to that in downstream) of filter is different in every filter stage. The difference depends on the size distribution of aerosols and the penetration curve of filter. We utilize this dependency of DF for estimation of the particle size distribution of aerosols. This method is not limited to radioactive aerosol. If appropriate quantitative analysis of collected sample is available, the method is applicable to aerosols in which interested from standpoints of industrial hygiene and public health.

In the present method, it is necessary for radioactivity of aerosol particle to be related to the particle size. In case of homogeneous aerosols generated from radioactive material in single chemical form, the radioactivity of one particle can be assumed to be proportional to a volume of the particle, namely, a cubic of the particle size. And it is also necessary for the penetration curve of the filter to be known before the size estimations by the present method.

DF of the single stage filter is defined by a ratio of the total radioactivity of particles in upstream of the filter to that of downstream particles.

$$DF = \frac{\int_0^{d_p} F(0,d_p) \cdot d(d_p) \int_0^{d_p} F(1,d_p) \cdot d(d_p)}{\int_0^{d_p} F(0,d_p) \cdot d(d_p)}$$  \hspace{1cm} (1)

where

- $d_p$: particle size,
- $F(0,d_p)$: particle size distribution function on radioactivity in upstream of the filter,
- $F(1,d_p)$: particle size distribution function on radioactivity in downstream of the filter.

When penetration function of the filter, $P(d_p)$, is given, $F(1,d_p)$ in Eq. (1) is replaced with $F(0,d_p) \cdot P(d_p)$.

$$DF = \int_0^{d_p} F(0,d_p) \cdot d(d_p) \int_0^{d_p} F(0,d_p) \cdot P(d_p) \cdot d(d_p)$$  \hspace{1cm} (2)

In the same manner, $DF(n)$ of $n$-th stage filter is defined by a ratio of the radioactivity of particles in upstream of $n$-th stage filter to that of downstream particles. Namely, $DF(n)$ is expressed by the following equation:

$$DF(n) = \frac{\int_0^{d_p} F(0,d_p) \cdot P(d_p)^{n-1} \cdot d(d_p)}{\int_0^{d_p} F(0,d_p) \cdot P(d_p)^n \cdot d(d_p)}$$  \hspace{1cm} (3)

On the other hand, an experimental decontamination factor is determined by measuring radioactivities on the filter. The experimental $DF$ of $n$-th stage filter is defined as a ratio of the upstream radioactivity of the filter to the downstream radioactivity. The upstream radioactivity is equal to the total radioactivity of particles collected on the $n$-th stage filter and the succeeding filters, $\sum_{i=n}^{m} A(i)$, where $A(i)$ is the measured radioactivity on the $i$-th stage filter. And the downstream radioactivity is equal to the total radioactivity of particles collected on the $(n+1)$-th stage filter and the succeeding filters, $\sum_{i=n+1}^{m} A(i)$. Namely, the experimental $DF$ is given by the following equation;

$$DF(n) = \frac{\sum_{i=n}^{m} A(i)}{\sum_{i=n+1}^{m} A(i)}$$  \hspace{1cm} (4)

If an absolute filter with 100% of particle collection efficiency is used as the $k$-th stage filter, infinities in Eq. (4) are replaced by $k$. For example, the experimental $DF$ of the 3rd stage filter in a 5 stage filter system is given by $(A(3) + A(4) + A(5)) / (A(4) + A(5))$.

The experimental $DF$ in Eq. (2) is easily obtained from radioactivity measurements of each filter. However, Eq. (3) is not mathematically reduced, even though $P(d_p)$ is known and is expressed as a function of $d_p$. Before size estimations, therefore, size distribution functions have to be assumed for all practically possible cases, and $DF$ values in Eq. (3) have to be calculated. And size distribution function of which calculated $DF$ is in accord with the experimental one, is to be selected among the calculated functions.

### III ANALYTICAL METHOD

#### 1. Preparations for data inversion

In order to make a numerical integration for Eq. (3) using a microcomputer, both the size distribution function and the penetration function should be given in a mathematical form. In this paper, the size distribution was assumed to be a log-normal distribution. Because there are many reports that most of size distributions of aerosols are close to a log-normal distribution. Consequently, the distribution is fixed by two independent variables, $MD$ (median diameter) and $GSD$ (geometric standard deviation). It means that only two $DF$ values of the first and the second
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stage filter are enough for estimations of size distribution.

For the penetration function of filter a parabola function on a log-log scaled graph of penetration vs. particle size was assumed in this paper. Because near parabolic curves are frequently observed for most filter although a theoretical inevitability does not exist. For instance, at the air flow rate of 1.33 cm/s the penetration curve of HE-40T filter (Toyo Roshi Co., Tokyo), which is widely used for a dust sampling in Japan, is found to have been given by the following equation in our previous study,\(^5\)

\[ Y = -2.25X^2 - 4.12X - 2.71 \]  
where \( X = \log(d_p) \), \( Y = \log(P) \).

From a viewpoint of filtration, the curve is characterized by the following three parameters; namely, the most penetrating particle size (MPPS), the maximum penetration (MP), and the full width at tenth maximum (FWTM). From the Eq. (5), the MPPS of the HE-40T filter is calculated to be 0.121 \( \mu m \) in diameter. The MP and the FWTM are 0.153 and 21.6, respectively.

Numerical integration of Eq. (3) was executed for particles in a size range from 0.001 to 100 \( \mu m \) by using a microcomputer (NEC PC-9801, Nippon Electric Co., Japan). In order to calculate DF values for practically possible size distributions, 29 MDs ranging from 0.1 to 10 \( \mu m \) and 28 GSDs from 1 to 5 were selected, which resulted 812 (\( = 29 \times 28 \)) sets of values were determined. As shown in Fig. 1, DF values for the HE-40T filter are ranged from under 10 to over 1,000. Since the penetration curve is symmetry as an axis where \( MD \) equals the MPPS, \( MD \) is the curves with a constant GSD is bivalent against DF, and it is theoretically impossible to distinguish between larger and smaller MDs. Compared with the DFs of the first stage filter, those of the second stage filter are always lower except when GSD is 1. The difference of the DFs becomes notable as GSD increased or \( MD \) goes away from the MPPS.

In order to observe the effect of penetration curve on DF values, three characterizing parameters, namely MPPS, MP and FWTM, were changed in a range from the half to the twice of the each original values. Examples of the effect calculated for the case of GSD to be 2 are shown in Fig. 2. The three parameters give the same changes to DF curves as they affect the penetration curves; i.e., the MPPS moves DF curves horizontally, and the MP does vertically. And the FWTM changes those curvature. This means that the MPPS affects an applicable particle size range and the MP does an applicable DF range and the FWTM does a resolution in estimation of size distribution. These information are very useful for the selection of filter media in the present method.

2. Inversion of size distribution from measured DF

Inversion method from the measured DF values to a size distribution is very simple, and it is only to select one combination with \( MD \) and GSD so that the theoretical DF agrees with the mea-
sured one. However, the measured DFs have always uncertainties resulted from a statistical fluctuation in a radiation counting, errors in measurements of a sampling air flow rate and so on. For practical purposes, therefore, all possible combinations of MD and GSD which correspond to the measured DF within the uncertainties should be firstly picked out from 812 distributions in every filter stage. Then only repeatedly picked combinations in each stage should be selected. The number of selected combinations is not always one, and it is dependent on a reliability of measured data and a propriety of assumptions made in calculation. Increase of filter stage can provide more information and lead to more precise size estimation.

When all the DFs in a set of filters calculated for a selected combination of MD and GSD are in accord with the measured DFs within the standard error, the size estimation is evaluated to be in the first level in accordance. In the case when one of the calculated DFs is outside the range, the estimation is in the second level. Thus all estimations are ranked to an appropriate level in order of the accordance.

IV EXPERIMENTAL VALIDATION OF THE MSF THEORY

In order to examine the propriety of the present MSF method, a comparison was made between the size distribution estimated by the MSF method and determined by a traditional method. For $^{198}$Au labeled gelatin aerosols, DF data were already obtained experimentally at the air flow rate of 1.33 cm/s in the three stage filter system.\(^5\) The DFs were measured to be 94.1±3.9, 20.7±1.1, and 13.9±2.1 for the filters of the first, the second and the third stage, respectively. And the size distribution of radioactive aerosols used in the DF measurements were directly determined to be AMAD = 1.2 μm and GSD = 2.0 by a cascade impactor method.

In the present MSF method, size distribution of radioactive aerosols is estimated from the experimentally measured DFs. It was first tried to select all size distributions having the same DFs with the measured DFs within the experimental errors. In the present example, however, no size distribution was selected in this first level of accordance. Therefore, the second level selection, in which only one DF is made an exception in the selection, was made, and three size distributions were selected. As shown in Table 1, the MDs of the selected size distributions were ranging from 0.9 to 1.2 μm and the GSDs were from 1.8 to 2.0. These three distributions were within a reasonably small range. And they were found to be very close to the distribution determined by the cascade impactor method.


Table 1  Comparison between the experimental \( DF \) and the calculated \( DF \).

<table>
<thead>
<tr>
<th>Filter</th>
<th>Experiment</th>
<th>Calculation</th>
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<tr>
<td></td>
<td>( MD = 1.2 ) (^{a}) ) ( GSD=2.0 )</td>
<td>Level 1(^{b})) ( MD =0.9 ) ( GSD=1.8 )</td>
</tr>
<tr>
<td>First stage</td>
<td>94.1 ± 3.9</td>
<td>89.6</td>
</tr>
<tr>
<td>Second stage</td>
<td>20.7 ± 1.1</td>
<td>21.4</td>
</tr>
<tr>
<td>Third stage</td>
<td>13.9 ± 3.1</td>
<td>13.1</td>
</tr>
</tbody>
</table>

\(^{a})\) Size distribution determined by a cascade impactor and \( DF \)s measured in the three stage filter \( (YAMADA \ et \ al., \ 1987) \). \(^{b})\) No size distribution was found in the first level.

V  CONCLUSION

A new method for the size estimation of radioactive aerosols was proposed and the propriety of the term was discussed. The method has the following advantages;

1. It needs only modification of a traditional dust sampler to a multistage filter sampler, so that the operation is very simple and the cost is very low.

2. The applicable particle size range and the accuracy can be changed by selecting a proper filter media and operational conditions.

3. Other quantitative analyses such as a chemical analysis can be applied.

At the same time, the method has some problems to be taken into account as follows;

1. Size measurements are not made for each aerosol particle. The method reveals only overall size, namely a size distribution with combination of \( MD \) and \( GSD \).

2. Penetration curve is a bivalent function mathematically, and no unique answer is given. In most of cases, however, health physicists can easily distinguish between true and false by circumstantial judgments. Furthermore, particle size which they are interested in for lung deposition is ranging from 0.2 to 10 \( \mu m \), where monovalent penetration curves like HE-40T filter in this paper can be found.

3. There are some assumptions in the present method. For example, particle size is assumed to be log-normally distributed. All size distributions are approximately fitted to be log-normal even though the distribution is bimodal or trimodal one. Theoretically, the present method can be applicable to these multimodal distribution by using more filter stages, but proper sampling with a five or six stage filter and their accurate measurement is very difficult practically.

These problems might be severe for the method to be used as an equipment for measurements in aerosol study. But in the field of radiological protection it is not always necessary to know the particle size distribution precisely. The accuracy of the measurement is not always considered to be the most important. We can conclude that the present method is simple enough to the practical air monitoring and the accuracy of the method is good enough to particle size measurements in the field of radiological protection.

REFERENCES