Journal of Environmental Protection, 2022, 13, 589-602
https://www.scirp.org/journal/jep
ISSN Online: 2152-2219
ISSN Print: 2152-2197

Meta-Study of Particulate Detection Losses on Radioactive Air Sample Filters

J. Matthew Barnett*, Hunter Z. Edwards

Pacific Northwest National Laboratory, Richland, USA
Email: *matthew.barnett@pnnl.gov

How to cite this paper: Barnett, J.M. and Edwards, H.Z. (2022) Meta-Study of Particulate Detection Losses on Radioactive Air Sample Filters. *Journal of Environmental Protection*, **13**, 589-602.
https://doi.org/10.4236/jep.2022.138037

Received: August 4, 2022
Accepted: August 23, 2022
Published: August 26, 2022

Abstract

Several mathematical relationships between air sample filter mass loading and the correlated analytical self-absorption factor were developed using data from other published research in this meta-study. Gross-alpha and -beta applications are addressed for this research. As filter media becomes loaded with particulate matter, there is potential for measurement losses due to self-absorption by mass loading. Components contributing to absorption include particulate dust, radioactive particulates, and filter material. Standards indicate a correction factor should be used when the penetration of radioactive material into the collection media or self-absorption of radiation by the material collected would reduce the detection rate by more than 5%. Previously, losses due to self-absorption have been reported up to 100% over a range up to ~10 $mg \cdot cm^{-2}$ mass loading. These absorption losses then can be used to determine a correction factor for sample results. For low mass loadings (e.g., ≤0.1 $mg \cdot cm^{-2}$) corrections factors in the 0.85 - 1 range have been recommended and used, while at higher mass loadings nearer to 10 $mg \cdot cm^{-2}$ correction factors closer to 0 (representing near 100% losses) are used. Based on data from published studies, the different methods for relating percent loss due to self-absorption to mass loading include linear, exponential, quadratic, and trinomial derived functions. Where applicable, both forced zero and non-forced zero results were evaluated. From the derived functions evaluated, the trinomial function provided the best fit. Once the sample filter mass loading is known, the trinomial function can be applied to estimate losses and the corresponding self-absorption factor. When applied to routine operating conditions for radiological facility stacks monitored at the Pacific Northwest National Laboratory for an average sample filter mass loading of 0.09 ± 0.12 (2σ) $mg \cdot cm^{-2}$ (excluding negative values and outliers) and a range from 0 - 0.24 $mg \cdot cm^{-2}$, the estimated trinomial function nominal self-absorption losses are less than 5% at 0.09 $mg \cdot cm^{-2}$ and less than 10% at 0.24 $mg \cdot cm^{-2}$. The trinomial function is one method that may be used to adjust the activity results of an air sample when the sample-specific mass loading is determined. The application of no correction factor when the ANSI/HPS N13.1-2021 guidance of a 5% threshold for loss is not reached with typical stack sample mass loadings may be reasonable in high-efficiency particulate air filtered systems. For simplicity, it would be conservative in assigning the self-absorption correction factor at the 5% threshold (*i.e.*, 0.95) for general uses but in cases of heavy mass loading to calculate the factor.

Keywords

Air Sampling, Environmental Radioactivity, Self-Absorption, Correction Factor, Standards

1. Introduction

Mathematical solutions are developed and evaluated in this meta-study from the data to determine the self-absorption factor of radioactive air sample filters. To perform monitoring of air emissions from laboratories that have the potential to emit radioactive particles, the Effluent Management group (EM) at Pacific Northwest National Laboratory (PNNL) coordinates the collection of particulate material from building emission stacks on 47-mm membrane filters. EM manages the analyses of the filters for gross alpha and gross beta activity as well as periodic composite isotope-specific analyses to determine the total radioactive air emissions. Only the gross alpha and gross beta measurements on sample filters are considered in this report. Guidance from American National Standards Institute (ANSI)/Health Physics Society (HPS) N13.1-2021, *Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stack and Ducts of Nuclear Facilities*, recommends that if the penetration of radioactive material into the filter collection media or self-absorption of radiation by the material collected would reduce the count rate of radioactive particles by more than 5%, a correction factor should be used [1]. Furthermore, correction factors may be applied to prevent the under-reporting of emissions. Since the mid-1980s, PNNL has used a 0.85 correction factor for the self-absorption of alpha particles based on the similarity of filter media, particle size, alpha energy (~5 MeV alpha energies [^{239}Pu]), and flow rates [2] [3]. To account for the activity that cannot be detected by direct counting of alpha or beta particles due to degradation or blocking by filter fibers or by inactive dust particles collected with the radioactive particles, EM has historically applied the same correction factor to samples analyzed for beta particles [4]. This 0.85 correction factor assumes approximately 15% losses in the count rate of both alpha and beta particles [5]. The self-absorption factor is different than the collection efficiency of the filter media itself; both factors, though, are generally applied to the reported sample emissions result. This meta-study was conducted to evaluate the 0.85 correction factor using published empirical data.

Since 1963, the effects of particle size and dust loading as it relates to alpha spectra on air sample filters have been reported [6]. Then in 1984, when D.P. Higby published the report, *Effects of Particle Size and Velocity on Burial Depth of Airborne Particles in Glass Fiber Filters*, it was accepted that absorption of alpha radiation emitted from airborne particles collected on glass-fiber filters does not constitute a major source of error in estimating concentrations of airborne alpha-emitting radionuclides [2]. In that report, Higby [2] evaluated the extent to which particle size and sampling velocity influence burial depth in glass-fiber filters. Aerosols of $^{239}PuO_2$ were collected and the fraction of counts lost due to burial was determined as the ratio of activity detected by direct alpha counting to the quantity determined by photon spectrometry. Results from Higby [2] indicate the minimum mass loading for a buried ^{239}Pu alpha particle to be undetected (100% loss) due to absorption is approximately 3.7 $mg \cdot cm^{-2}$ based on the degradation of the alpha energy spectrum (e.g., range) associated with the depth of the particle in the filter material. In other words, at least 3.7 $mg \cdot cm^{-2}$ is required to fail to detect a representative alpha particle and not that 100% losses would be expected at that value. The Higby [2] study 1) reports that the burial of airborne particles collected on glass-fiber filters appears to be a weak function of sampling velocity and particle size within the ranges studied, 2) recognizes that glass-fiber filters tend to be surface loading, thereby reducing the need for correction for burial losses, and 3) does not consider additional dust loading from non-radioactive material. The results indicate an assumed 10% - 15% loss (0.85 - 0.90 correction factor) would make sure concentrations of airborne alpha-emitting radionuclides would not be underestimated by collection and analysis on glass-fiber filters.

This meta-study derives a relationship between filter mass loading and percent loss using results from previous studies of self-absorption and assumes that the vast majority of the particulate loading rests on the surface of the filter fiber media. Mass loading from routine operations consists of particulate dust and radioactive particulates collected on the filter. Radioactive material is considered to be uniformly distributed across the active area of the filter and not just on the top layer, and the mass loading should not include the full depth of the filter material. By exploring other research studies, a more comprehensive representation of the percent loss of alpha particles due to mass loading on sample filters rather than just burial depth is compiled and compared to the previously accepted correction factor of 0.85 while noting a correction factor of 1 would indicate that no correction is needed.

These terms are defined here to provide consistency between the reports in this meta-study. Mass loading consists of the particulate dust and radioactive particulates collected on the filter material; the filter material is excluded for the purposes of determining mass loading. Mass loading is typically reported in units of mass per active area of the sample filter. Self-absorption occurs when the emitted radiation does not reach the detector because of interference from other particulate matter on the filter or the filter media. Self-absorption is accounted for by applying a correction factor to the sample result; the correction factor generally is applied equally to both alpha and beta counting results. Additional information and equations for applying correction factors, including the self-absorption correction factor, can be found in Barnett [4]. For clarity, the self-absorption correction factor in decimal format is a divisor applied to correct the sample data. Correction factors in this meta-study are in a consistent format and may have been converted from their original form. The percent loss, in decimal format, is one minus the self-absorption correction factor (e.g., 15% loss corresponds to a self-absorption correction factor of 0.85).

The mass loading aspect of each report in this meta-study is summarized below. They describe our investigations of data presented in Luetzelschwab *et al.* [7], Huang *et al.* [8], Barnett *et al.* [9], Smith *et al.* [5], and Hogue *et al.* [10]. Losses of 100% are not considered because there is an expectation that some radioactive material (*i.e.*, the top thin layer) will always be counted on a sample filter regardless of the total mass loading. It is recognized the measurement of radioactivity from aerosol deposited on/in an air sample filter is complicated and that the studies included here had disparate methods, goals, techniques and end points. However, these studies are combined here, using the salient information available, such that a mathematical relationship between correction factors and mass loading could be formed.

Luetzelschwab *et al.* [7] utilized bi-layer fiberglass filters with sample particles collected from the air. Counting efficiencies were reported as a result of the absorber thickness. The areal density of the front layer of the filter is reported and can therefore be separated from the reported results. Mass loadings in the study included dust loading plus the mass of the filter front layer and are less when the filter front layer is eliminated. Luetzelschwab *et al.* [7] reports that depending on the type of filter used, dust loading on the filter may not impair the sample results provided the deposited layer remains thin (≤0.1 $\text{mg}\cdot\text{cm}^{-2}$). Data included from Luetzelschwab *et al.* [7] indicate that for a mass loading, which includes the areal density of the front layer of the filter, of 2.3 $\text{mg}\cdot\text{cm}^{-2}$, the calculated loss of alpha particles is 28%. Also included in this study is a reported 40% loss when a mass loading of 3.3 $\text{mg}\cdot\text{cm}^{-2}$ is present. When the areal density of the front layer of the filter (*i.e.*, 1.3 $\text{mg}\cdot\text{cm}^{-2}$) is removed, the losses are 28% and 40% for mass loadings of 1 $\text{mg}\cdot\text{cm}^{-2}$ and 2 $\text{mg}\cdot\text{cm}^{-2}$, respectively.

Huang *et al.* [8] used three types of membrane filters in this study in which suspended particles were deposited by dry pneumatic dispersion. Thin layer deposits of radioactive aerosols were not significantly degraded by an underlying thick layer of dust and the filter matrix was excluded. They reported the thickness of an underlying dust layer did not show a significant effect for the materials studied and a loading range of 0.01 to 10 $\text{mg}\cdot\text{cm}^{-2}$. The detection of radioactive aerosols with little deterioration in energy resolution is possible if the deposited layer of radioactive aerosols remains thin (≤0.1 $\text{mg}\cdot\text{cm}^{-2}$).

Barnett *et al.* [9] used acrylic copolymer filters on a nylon substrate in which particles were collected from the ventilated (stack) exhaust stream in nuclear facilities. Examination showed the majority of particles were found to remain on the surface of the filter. Results indicate that at low mass loadings (0.08 ± 0.05 mg·cm^{-2}) more activity is measured by directly counting the sample filter rather than by counting after an acid digestion process with a wide variation encompassing the value of 1 where no difference can be detected between the two separate count processes.

Smith *et al.* [5] is a follow-on study from Barnett *et al.* [9] and also used acrylic copolymer filters on a nylon substrate. Particles were collected from stack exhaust streams. Smith *et al.* [5] note the acid-digestion process should have eliminated the effects of self-absorption or penetration into the collection media, but for most samples, the values of after-digestion results were lower than those measured before digestion. This finding is attributed to the lack of precision in the digestion, analytical process, and large instrument sensitivity error at the extremely low levels of radioactivity on the air sample filters. Smith *et al.* [5] report the results of mass loading based on the weighing before and after installation of air sample filters with a range of 0 mg·cm^{-2} to 0.24 mg·cm^{-2}. Using results previously reported by Higby [2] and Luetzelschwab *et al.* [7], both a linear fit and an exponential fit to the data were derived.

Hogue *et al.* [10] developed correction factors based on sampled activity to air volume followed by Monte Carlo modeling. They used glass fiber filters and particles were collected from occupational airborne radioactivity monitoring. The filter matrix was excluded from the analysis, and a tiered approach for correction factor principles was developed. Samples in the developed dataset appeared to have a maximum loading of about 9 mg·cm^{-2}. The correction factors do not have a linear relationship with dust loading, and the reported approach is used and applied for varying mass loading quantities.

For this meta-study, the dust loading, which includes the radioactive material, on the filter is roughly expected to be uniformly dispersed throughout the dust. Sampling is a continuous process where additional sample material continues to accumulate on the top surface. Accident or abnormal conditions resulting in a one-time "poof" of radioactivity are not considered, and a one-time release later covered by additional dust would potentially yield a very different result than the chronic activity interspersed by dust presented here. Hence, the mass loading (mg·cm^{-2}) is the important factor considered, and it does not directly take into account interference from the filter medium due to the impact depth.

2. Methods and Data

In total, information from the five research studies described above was combined and evaluated in this meta-study to develop unique functions of mass loading that then could be used to determine a self-absorption correction factor to sample data. Each study revealed fewer detectable particles with increasing mass loading on the filter. **Table 1** shows the complete list of mass loadings and associated percent losses and correction factors.

Table 1. List of mass loadings and associated percent losses and self-absorption correction factors.

Mass Loading (mg/cm^2)	Reported % Loss	Self-Absorption Correction Factor	Source
10	99	0.01	Assumed upper bound
9	77	0.23	Hogue *et al.* [10]
8.5	75	0.25	Hogue *et al.* [10]
8	74	0.26	Hogue *et al.* [10]
7.5	73	0.27	Hogue *et al.* [10]
7	71	0.29	Hogue *et al.* [10]
6.5	70	0.30	Hogue *et al.* [10]
6	68	0.32	Hogue *et al.* [10]
5.5	66	0.34	Hogue *et al.* [10]
5	63	0.37	Hogue *et al.* [10]
4.5	60	0.40	Hogue *et al.* [10]
4	57	0.43	Hogue *et al.* [10]
3.5	53	0.47	Hogue *et al.* [10]
3.07	79	0.21	Smith *et al.* [5]
3	48	0.52	Hogue *et al.* [10]
2.5	42	0.58	Hogue *et al.* [10]
2	35	0.65	Hogue *et al.* [10]
2	40	0.60	Luetzelschwab *et al.* [7]
1.71	44	0.56	Smith *et al.* [5]
1.19	30	0.70	Smith *et al.* [5]
1	28	0.72	Luetzelschwab *et al.* [7]
0.66	17	0.83	Smith *et al.* [5]
0.24	6	0.94	Smith *et al.* [5]
0.13	0.01[a]	1.00	Barnett *et al.* [9]
0.01	0.01[a]	1.00	Huang *et al.* [8]
0.1	5	0.95	Huang *et al.* [8]
0.9	29	0.71	Hogue *et al.* [10]
0.05	17	0.83	Hogue *et al.* [10]
0.09	2	0.98	Smith *et al.* [5]
1.0E-4	0.01[a]	1.00	New filter

[a] Near-zero value to represent no observed self-absorption.

Luetzelschwab *et al.* [7] reported that when the front layer areal density of 1.3 $\text{mg}\cdot\text{cm}^{-2}$ is removed, the losses are 28% and 40% for mass loadings of 1 $\text{mg}\cdot\text{cm}^{-2}$ and 2 $\text{mg}\cdot\text{cm}^{-2}$, respectively. This results in correction factors of 0.72 and 0.60 for the mass loadings of 1 $\text{mg}\cdot\text{cm}^{-2}$ and 2 $\text{mg}\cdot\text{cm}^{-2}$, respectively.

Huang *et al.* [8] reported that the detection of radioactive aerosols with little deterioration in energy resolution is possible provided the deposited layer of radioactive aerosols remains thin. They also reported that the thickness of an underlying dust layer did not show a significant self-absorption when the deposited layer remained ≤ 0.1 $\text{mg}\cdot\text{cm}^{-2}$ and the loading range was 0.01 - 10 $\text{mg}\cdot\text{cm}^{-2}$. An assumption then is made regarding mass loading of the Huang *et al.* [8] results to assign 5% loss at 0.1 $\text{mg}\cdot\text{cm}^{-2}$ based on the recommendation of ANSI/HPS N13.1-2021 [1], and ~0% loss at 0.01 $\text{mg}\cdot\text{cm}^{-2}$. The resultant correction factors are 0.95 and 1 for mass loadings of 0.1 $\text{mg}\cdot\text{cm}^{-2}$ and 0.01 $\text{mg}\cdot\text{cm}^{-2}$, respectively.

Barnett *et al.* [9] indicate there is generally no observed activity difference between counting a sample filter directly and counting it again after acid digestion. Using the reported upper range, losses are expected to be ~0% for mass loadings ≤ 0.13 $\text{mg}\cdot\text{cm}^{-2}$, and a correction factor of 1 could be applied (*i.e.*, there is no indicated need for a correction factor to be applied).

Smith *et al.* [5] reported typical samples losses for both exponential and linear relationships and for typical average mass loadings of 0.09 $\text{mg}\cdot\text{cm}^{-2}$ show losses of approximately 2% with a linear model and 17% with an exponential model. Their data is dependent on Higby [2] and Luetzelschwab *et al.* [7]. This meta-study only utilized the linear model results rather than the exponential model results because the linear model results were consistent with other reports and had a zero intercept. Normal operating conditions show a range of mass loading results from 0 to 0.24 $\text{mg}\cdot\text{cm}^{-2}$, and there are four mass loading results greater than 0.24 $\text{mg}\cdot\text{cm}^{-2}$ that they considered being atypical. Losses of approximately 6% for the linear model were determined at 0.24 $\text{mg}\cdot\text{cm}^{-2}$. Percent losses for the linear model for the four reported atypical mass loadings are also included in the dataset in **Table 1**. The resultant correction factors for typical average mass loadings of 0.09 $\text{mg}\cdot\text{cm}^{-2}$ are 0.98 for the linear model.

According to Hogue *et al.* [10], the following approach for correction factor principles apply. A range of correction factors are generated and applied for varying mass loading quantities.

1) At dust loading levels less than 0.1 $\text{mg}\cdot\text{cm}^{-2}$, the loss is 17% and the correction factor is 0.83.

2) At dust loading levels equal to or greater than 0.1 $\text{mg}\cdot\text{cm}^{-2}$ but less than 1.7 $\text{mg}\cdot\text{cm}^{-2}$, the loss is 29% and the correction factor is 0.71.

3) At dust loading levels between 1.7 $\text{mg}\cdot\text{cm}^{-2}$ to 9 $\text{mg}\cdot\text{cm}^{-2}$, the losses range between 29% and 77%, respectively. The correction factor is given by $(0.744 + 0.39255\text{' dust loading})^{-1}$ where the dust loading is in $\text{mg}\cdot\text{cm}^{-2}$. The correction factor ranges from 0.71 to 0.23.

4) For samples with air volumes > 1024 m^3 (or assumed mass loadings > 9

mg·cm^{-2}), the losses are greater than 77%. The correction factor is less than 0.23.

By using these respective datasets, a master list of mass loadings and associated percent loss (% Loss) and self-absorption correction factors were compiled and labeled with each credited research report (**Table 1**). Although 100% self-absorption is not expected because the thin layer of radioactive material on the filter surface can always be counted and although values greater than 9.0 mg·cm^{-2} have not been investigated, an upper bound mass loading of 10 mg·cm^{-2} was assumed for this meta-study to have 99% losses and a correction factor of 0.01. This upper bound could just as easily be >10 mg·cm^{-2} but was chosen because of its near proximity to the overall reported results.

3. Results

Data from **Table 1** were then graphed using Microsoft Excel (Microsoft, One Microsoft Way, Redmond, Washington 98052 USA) to obtain alternative mathematical functions for linear, exponential, quadratic, and trinomial approaches. Data were evaluated with and without a forced zero intercept. The R^2 values were determined for each derived function. **Table 2** shows the equations and R^2 values for each mathematical function developed and considered. Plots generated, as seen in **Figures 1-6**, were for the mass loading versus percent losses from **Table 1** for each of the derived functions shown in **Table 2**. Exponential function results are not discussed further due to the demonstrated poor fit to the data (R^2 = 0.46). The y-axis represents the percent loss due to self-absorption, and the x-axis indicates mass loading in mg·cm^{-2}. **Table 3** shows various mass loadings and the resultant percent loss for the various functions.

For the forced zero intercepts of the derived functions, it is assumed that at 0 mg·cm^{-2} (*i.e.*, a new, unused sample filter), no losses occur. However, for the functions without a forced zero intercept, calculated losses range from 2% for the trinomial function to ~15% for the linear function. The high losses at 0 mg·cm^{-2} for the linear function corroborate the non-linear nature of data reported in Hogue *et al.* [10].

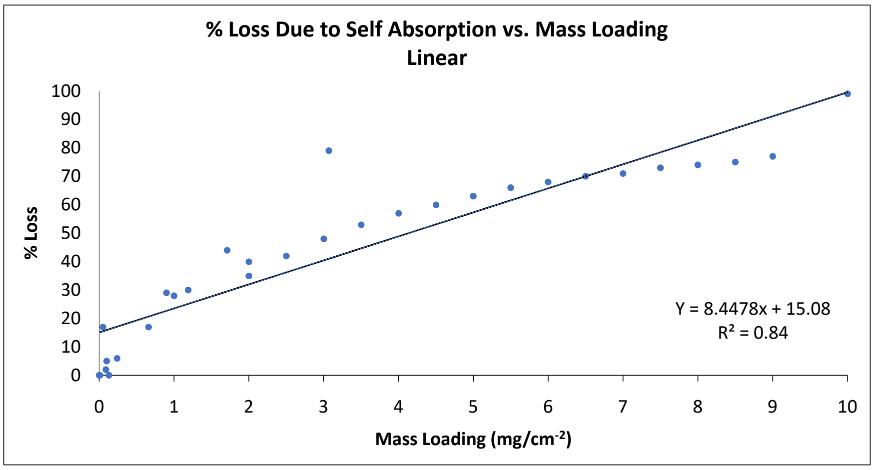

Figure 1. Linear function showing percent loss due to self-absorption vs. mass loading.

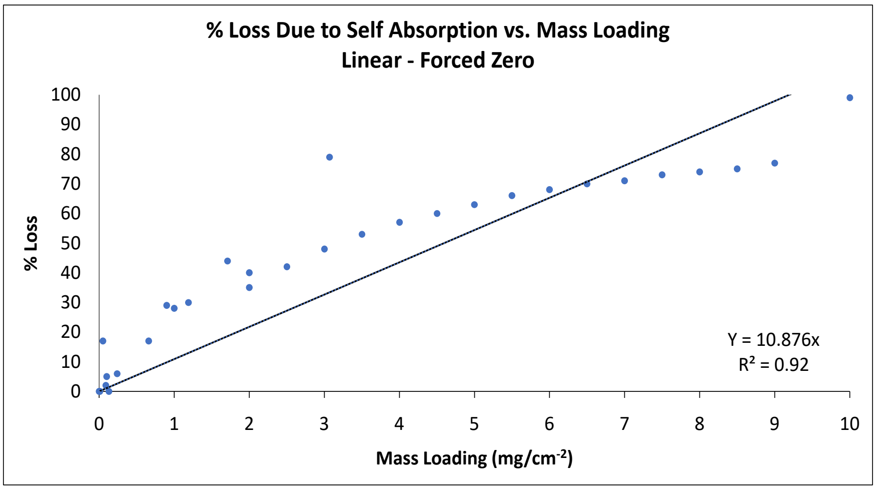

Figure 2. Forced zero intercept linear function showing percent loss due to self-absorption vs. mass loading.

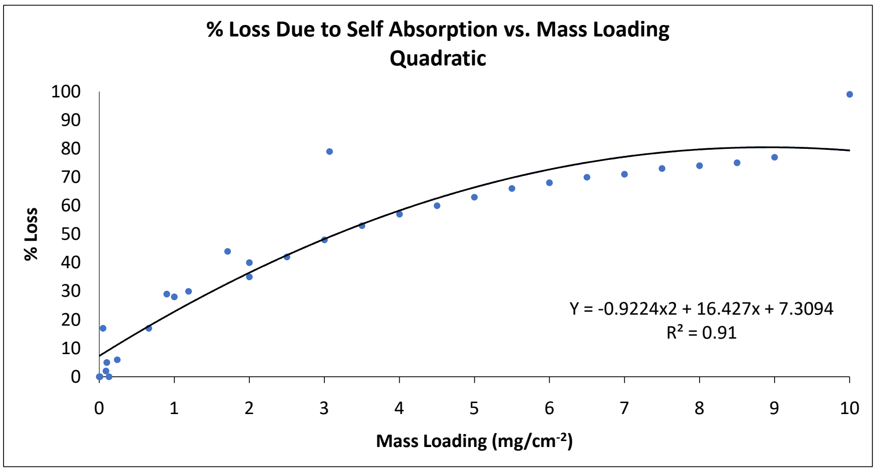

Figure 3. Quadratic function showing percent loss due to self-absorption vs. mass loading.

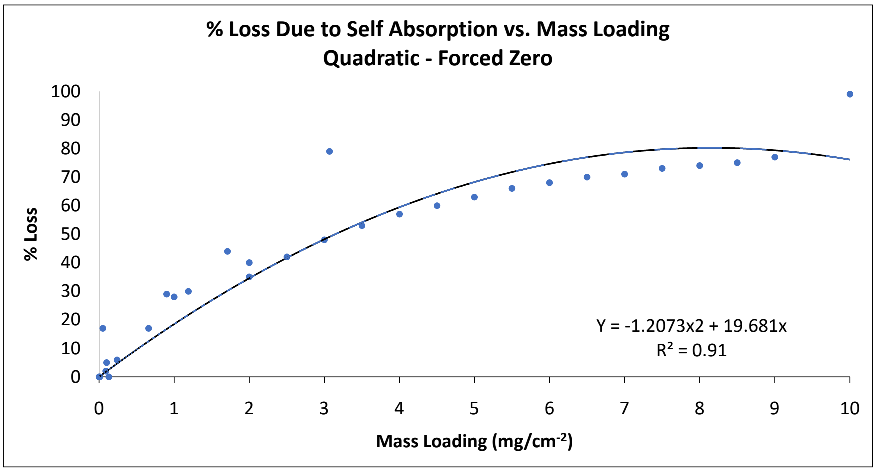

Figure 4. Forced zero intercept quadratic function showing percent loss due to self-absorption vs. mass loading.

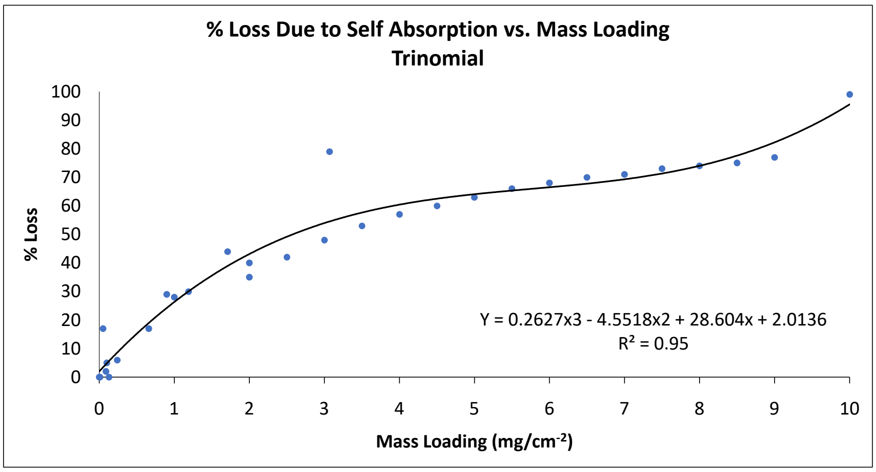

Figure 5. Trinomial function showing percent loss due to self-absorption vs. mass loading.

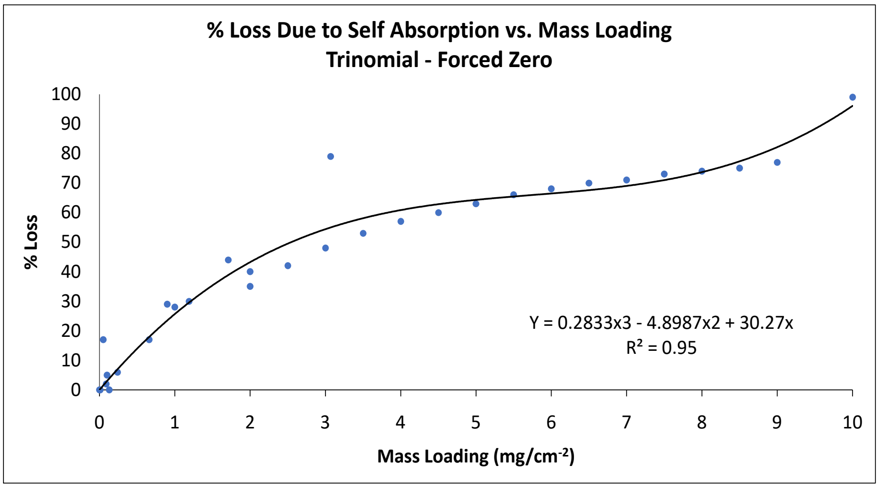

Figure 6. Forced zero intercept trinomial function showing percent loss due to self-absorption vs. mass loading.

Table 2. Derived mathematical functions for calculating the self-absorption percent loss from filter mass loading.

Graph Type	Function Derived[a]	R^2
Linear	$Y = 8.4478x + 15.08$	0.84
Linear (Forced Zero Intercept)	$Y = 10.876x$	0.92
Exponential[b]	$Y = 3.0094e^{0.4923x}$	0.46
Quadratic	$Y = -0.9224x^2 + 16.427x + 7.3094$	0.91
Quadratic (Forced Zero Intercept)	$Y = -1.2073x^2 + 19.681x$	0.91
Trinomial	$Y = 0.2627x^3 - 4.5518x^2 + 28.604x + 2.0136$	0.95
Trinomial (Forced Zero Intercept)	$Y = 0.2833x^3 - 4.8987x^2 + 30.27x$	0.95

[a] Where Y = self-absorption percent loss, and x = mass loading in $\text{mg}\cdot\text{cm}^{-2}$. [b] Forced zero intercept not possible for the exponential function.

Table 3. Comparison of various mass loads versus calculated percent loss.

Mass Loading (mg/cm^2)	Linear (% Loss)	Linear Forced Zero (% Loss)	Quadratic (% Loss)	Quadratic Forced Zero (% Loss)	Trinomial (% Loss)	Trinomial Forced Zero (% Loss)
0	15.1	0.0	7.3	0.0	2.0	0.0
0.1	15.9	1.1	8.9	2.0	4.8	3.0
0.2	16.8	2.2	10.6	3.9	7.6	5.9
0.3	17.6	3.3	12.2	5.8	10.2	8.6
0.4	18.5	4.4	13.7	7.7	12.7	11.3
0.5	19.3	5.4	15.3	9.5	15.2	13.9
1	23.5	10.9	22.8	18.5	26.3	25.7
2	32.0	21.8	36.5	34.5	43.1	43.2
3	40.4	32.6	48.3	48.2	54.0	54.4
5	57.3	54.4	66.4	68.2	64.1	64.3
7	74.2	76.1	77.1	78.6	69.3	69.0
10	99.6	100+	79.3	76.1	95.6	96.1

Various mass loadings were applied to the resultant functions to evaluate the recommendation of a 5% limit for self-absorption losses (*i.e.*, a correction factor of 0.95) in the ANSI/HPS N13.1-2021 [1] standard and the PNNL historically applied 15% self-absorption (correction factor of 0.85). The calculated percent losses of 5%, 15%, and maximum losses up to 100% are shown in **Table 4** for the mass loading associated with each function. At losses of 5%, the mass loading range is up to 0.46 $mg \cdot cm^{-2}$ (linear forced-zero function), and for losses of 15%, the mass loading range is up to 1.38 $mg \cdot cm^{-2}$ (linear forced-zero function). The quadratic forced zero, trinomial, and trinomial forced zero functions have 5% losses in the reported typical mass loading range of ~0.1 $mg \cdot cm^{-2}$ and up to ~0.24 $mg \cdot cm^{-2}$ reported by Smith *et al.* [5] and Barnett *et al.* [9]. The range of mass loadings resulting in 15% losses are 0.48 - 1.38 $mg \cdot cm^{-2}$ and in the approximate range of the four atypical mass loading results reported by Smith *et al.* [5].

Modeling indicates maximum losses for mass loadings > 8 $mg \cdot cm^{-2}$ for the quadratic and quadratic forced zero functions. An interesting aspect of the quadratic functions is that maximum losses are limited to about 80%. The linear and trinomial functions tend to be 9 - 11 $mg \cdot cm^{-2}$ for 100% losses, which also fits the expected upper bound of Hogue *et al.* [10].

The functions derived in this meta-study resulted in R^2 values in the 0.46 - 0.95 range. As previously noted, the data are not considered to be linear, and the R^2 values are 0.84 for the linear function and 0.92 with a linear forced zero intercept. As discussed earlier, the exponential function has the lowest R^2 value which indicates it is not a very good fit for the data. While the quadratic functions have R^2 values of 0.91, these functions are parabolic and do not continue to increase as the mass loading increases. Finally, the trinomial functions both have R^2 values of 0.95 which is the highest of the function groupings and the least disparity in percent losses between the derived function versus the forced-zero derived function. The trinomial functions appear best at bringing together the various aspects of the Hogue *et al.* [10] approach.

Using only trinomial functions as best-fit results in an intercept of 2.0% without a forced-zero intercept. **Table 5** shows the resultant trinomial functions for various expected losses at nominal, 5%, 15%, and 100% mass loadings. The nominal mass loading range of 0.09 mg·cm^{-2} shows the expected losses are ~2% - 5% while the 0.24 mg·cm^{-2} upper end of typical mass loadings shows expected losses of ~7% - 9%. At about 0.50 mg·cm^{-2} the expected losses are ~15% which would be indicative of the infrequent atypical sample mass loading results at PNNL.

Table 4. Comparison of various mass loads and calculated percent losses for 5%, 15%, and 100%.

Mass Loading (mg/cm^2)	Linear[a] (% Loss)	Linear Forced Zero (% Loss)	Quadratic[b, c] (% Loss)	Quadratic Forced Zero[c] (% Loss)	Trinomial (% Loss)	Trinomial Forced Zero (% Loss)
0.11					**5**	
0.17						**5**
0.26				**5**		
0.46		**5**				
0.48			**15**			
0.49					**15**	
0.54						**15**
0.80				**15**		
1.38		**15**				
8.20				**80**		
8.90			**80**			
9.20		**100**				
10.05	**100**					
10.21						**100**
10.26					**100**	

[a]The Linear function is greater than 15% at 0 mg cm^{-2}. [b]The Quadratic function is greater than 5% at 0 mg cm^{-2}. [c]The Quadratic and Quadratic Forced Zero functions show the maximum percent loss which is less than 100%.

Table 5. Final derived trinomial functions for calculating the self-absorption percent loss from filter mass loading.

Mass Loading (x)	Trinomial Function $Y = 0.2627x^3 - 4.5518x^2 + 28.604x + 2.0136$		Trinomial Function Forced Zero $Y = 0.2833x^3 - 4.8987x^2 + 30.27x$	
(mg/cm^2)	(% Loss)	(Correction Factor)	(% Loss)	(Correction Factor)
0.09[a]	4.6	0.954	2.7	0.973
0.11	**5.0**	**0.950**	3.1	0.969
0.17	6.7	0.933	**5.0**	**0.950**
0.24[b]	8.6	0.914	7.0	0.930
0.49	**15.0**	**0.850**	13.7	0.863
0.54	16.2	0.838	**15.0**	**0.850**
10.21	99.2	0.008	**100.0**	-
10.26	**100.0**	-	100+	-

[a]Nominal average sample filter mass loading at PNNL [5]. [b]Upper range limit of sample filter mass loading at PNNL [5].

4. Conclusions

Correction factors are applied commonly to address measurement errors, prevent under-reporting of emissions, and conservatively report emissions [4]. This meta-study evaluated linear, exponential, quadratic, and trinomial derived mathematical functions that relate percent loss due to self-absorption associated with filter mass loading. The percent loss can then be used to determine a correction factor for self-absorption.

In reviewing the derived functions (Table 2) and the resultant percent losses (Tables 3-5), a forced-zero fit represents results associated with a new unused filter, while the non-forced zero functions give an intercept approximating an estimated minimum self-absorption factor (e.g., perhaps due to impaction depth considerations). The trinomial forced zero function is recommended for routine samples with normal mass loadings to determine percent loss and associated self-absorption correction factors. It is particularly useful when the mass loading is known. Using this approach, the ANSI/HPS N13.1-2021 [1] standard may be satisfied by applying a nominal 5% (or 0.95 self-absorption correction factor) to typical PNNL sampling activities with known mass loadings of up to 0.24 $mg \cdot cm^{-2}$.

It is important to understand the overall mass loading on the sample filter and then apply an appropriate correction factor. A static factor of 0.95 could generally account for the small variations associated with self-absorption while not underestimating annual emissions results. For heavier mass loading situations, say greater than 1.0 $mg \cdot cm^{-2}$, the trinomial function with forced zero intercept may yield a better overall result in determining the losses. Nevertheless, users involved in radioactive air monitoring activities should work to develop a self-absorption correction factor that can be defended whether by use of this model, another model, or actual research results.

Acknowledgements

This work was conducted at the Pacific Northwest National Laboratory, which is operated for the U.S. Department of Energy by Battelle under Contract DE AC05 76RL01830.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Health Physics Society (2021) Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities. Health Physics Society, Herndon.

[2] Higby, D.P. (1984) Effects of Particle Size and Velocity on Burial Depth of Airborne Particles in Glass Fiber Filters. Pacific Northwest Laboratory, Richland. https://doi.org/10.2172/6312811

[3] Barnett, J.M., Cullinan, V.I., Barnett, D.S., Trang-Le, T.L.T., Bliss, M., Greenwood, L.R. and Ballinger, M.Y. (2009) Results of Self-Absorption Study on the Versapor 3000 Filters for Radioactive Particulate Air Sampling. Pacific Northwest National Laboratory, Richland. https://doi.org/10.2172/949086

[4] Barnett, J.M. (2011) Concepts for Environmental Radioactive Air Sampling and Monitoring. In: Ekundayo, E.O. Ed., *Environmental Monitoring*, IntechOpen, Rijeka. https://doi.org/10.5772/28533

[5] Smith, B.M., Barnett, J.M. and Ballinger, M.Y. (2011) Assessment of the Losses due to Self-Absorption by Mass Loading on a Radioactive Particulate Air Stack Sample Filters. Pacific Northwest National Laboratory, Richland. https://doi.org/10.2172/1004502

[6] Stevens, D.C. and Toureau, A.E.R. (1963) The Effect of Particle Size and Dust Loading on the Shape of Alpha Pulse-Height Spectra of Air Sample Filters. Atomic Energy Research Establishment, Harwell.

[7] Luetzelschwab, J.W., Storey, C., Zraly, K. and Dussinger, D. (2000) Self-Absorption of Alpha and Beta Particles in a Fiberglass Filter. *Health Physics*, **79**, 425-430. https://doi.org/10.1097/00004032-200010000-00012

[8] Huang, S., Schery, S.D., Alcantara, R.E., Rodgers, J.C. and Wasiolek, P.T. (2002) Influence of Dust Loading on the Alpha-Particle Energy Resolution of Continuous Air Monitors for Thin Deposits of Radioactive Aerosols. *Health Physics*, **83**, 884-891. https://doi.org/10.1097/00004032-200212000-00016

[9] Barnett, J.M., Cullinan, V.I., Barnett, D.S., Trang-Le, T.L.T., Bliss, M., Greenwood, L.R. and Ballinger, M.Y. (2009) Results of Self-Absorption Study on the Versapor 3000 47-mm Filters for Radioactive Particulate Air Stack Sampling. *Health Physics*, **97**, S161-S168. https://doi.org/10.1097/HP.0b013e3181b2713e

[10] Hogue, M.G., Gause-Lott, S.M., Owensby, B.N., Slack, T.M., Smiley, J.J. and Burkett, J.L. (2018) Alpha Air Sample Counting Efficiency versus Dust Loading: Evaluation of a Large Data Set. *Health Physics*, **114**, 479-485. https://doi.org/10.1097/HP.0000000000000800