Pacific Northwest NATIONAL LABORATORY

PNNL-30964

Collective Analysis of Alpha Particle Losses Due to Self-Absorption by Mass Loading on Radioactive Particulate Glass Fiber Filters

February 2021

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JM Barnett

U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
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Summary

In this study, we derived a relationship between filter mass loading and the percent loss during analysis using the mass loading data collected from six previous studies of self-absorption. Components of mass loading include particulate dust, radioactive particulates, and filter material. In a research report published in 1984, Higby[1] calculated a minimum burial depth for an alpha particle to be lost due to absorption (100% loss) of about 3.7 mg/cm^2 based on calculations for the range of 239-Pu alpha particles in glass fiber filters. From there, Higby[1] concluded that a correction factor of 0.85 assumes approximately 15% losses in the count rate of both alpha and beta particles. In 2000, Luetzelschwab et al.[2] recommended assuming a 40% loss at a loading of 3.3 mg/cm^2 and a 28% loss for a loading of 2.3 mg/cm^2 which included the frontal face mass of the filter. More recently, the 100% losses due to absorption were reported to be in the 10 mg/cm^2 range.[3,4] Presented here is a trinomial relationship method of relating percent loss due to self-absorption to filter mass loading, based on data reported by Higby,[1] Luetzelschwab et al.,[1] Huang et al.,[1] Hogue et al.[1], Barnett et al.,[5] and Smith et al.[6]

Under normal operating conditions at the stacks monitored by Effluent Management, the mass loading of sample filters averages 0.09 ± 0.12 (2σ) mg/cm^2 (excluding negative values and outliers) and ranges from 0 mg/cm^2 to 0.24 mg/cm^2.[5, 6] Based on current mass loading results for Effluent Management stack sample filters, the forced-zero trinomial relationship method estimated self-absorption losses of less than 5%. Because American National Standards Institute/Health Physics Society N13.1-2011[7] guidelines 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 count rate by more than 5%, it is possible continued application of a correction factor to the Effluent Management stack samples is no longer necessary. Nevertheless, continuing to assign a correction factor at the 5% threshold (i.e., 0.95) would be a conservative approach.

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[1] Higby DP. 1984. *Effects of Particle Size and Velocity on Burial Depth of Airborne Particles in Glass Fiber Filters*. PNL-5278, Pacific Northwest Laboratory, Richland, Washington.
[2] Luetzelschwab JW, C Storey, K Zraly, and D Dussinger. 2000. “Self-absorption of Alpha and Beta Particles in a Fiberglass Filter.” *Health Physics* 79(4):425–430.
[3] Huang S, SD Schery, RE Alcantara, JC Rodgers, and PT Wasiolek. 2002. “Influence of Dust Loading on the Alpha-Particle Energy Resolution of Continuous Air Monitors for Thin Deposits of Radioactive Aerosols.” *Health Physics* 83(6):884–891.
[4] Hogue MG, SM Gause-Lott, BN Owensby, TM Slack, JJ Smiley, and JL Burkett. 2018. “Alpha Air Sample Counting Efficiency Versus Dust Loading: Evaluation of a Large Data Set.” *Health Physics* 114(5):479–485.
[5] Barnett JM, VI Cullinan, DS Barnett, TLT Trang-Le, M Bliss, LR Greenwood, and MY Ballinger. 2009b. “Results of Self-Absorption Study on the Versapor 3000® 47-mm Filters for Radioactive Particulate Air Stack Sampling.” *Health Physics-Operational Radiation Safety* 97(5):S161–S168.
[6] Smith BM, JM Barnett, and MY Ballinger. 2011. *Assessment of the Losses Due to Self Absorption by Mass Loading on a Radioactive Particulate Air Stack Sample Filters*. PNNL-20098, Pacific Northwest National Laboratory, Richland, Washington.
[7] American National Standards Institute. 2011. *Sampling and Monitoring Releases of Airborne Radioactive Substances From the Stacks and Ducts of Nuclear Facilities*. Health Physics Society, ANSI/HPS N13.1-2011, McLean, Virginia.

Acknowledgments

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Acronyms and Abbreviations

ANSI	American National Standards Institute
DOE	U.S. Department of Energy
EM	Effluent Management group
HPS	Health Physics Society
PNNL	Pacific Northwest National Laboratory
SAF	self-absorption factor

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1.0 Introduction

To perform environmental monitoring of air emissions from laboratories that have the potential to emit radioactive particles, the Effluent Management group (EM) of Pacific Northwest National Laboratory (PNNL) coordinates the collection of particulate material from building emission stacks on 47 mm Versapor® 3000 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 amount of 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) N13.1-2011, *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 (HPS 2011). Furthermore, correction factors are commonly applied to prevent under-reporting of emissions and to maintain a conservative emission result (Barnett 2011). Since the mid-1980s, PNNL has used a 0.85 correction factor for self-absorption of alpha particles based on similarity of filter media, particle size, and flow rates (Higby 1984, Barnett et al. 2009a). EM has historically applied the same correction factor equally to samples analyzed for beta particles. This 0.85 correction factor assumes approximately 15% losses in the count rate of both alpha and beta particles (Smith et al. 2011). 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.

Since 1963, the effects of particle size and dust loading as it relates to alpha spectra on air sample filters has been reported (Stevens and Toureau 1963). 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 after the fact 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. Higby suggests the minimum burial depth for an alpha particle to be lost (100% loss) due to absorption is "… approximately 0.0037 g/cm^2, or 3.7 mg/cm^2." This report investigates this minimum burial depth value to determine if sample filters have a higher minimum burial depth for alpha particles to be lost, such as at or near 10 mg/cm^2. By exploring other, more recent research studies such as Hogue et al. (2018), Huang et al. (2002), and others, the relationship of the data sets, when compiled, suggest a more accurate representation of percent loss of alpha particles due to mass loading on sample filters rather than just burial depth. The newer data also suggests that the previously accepted correction factor of 0.85 should be revisited and updated where a correction factor of 1 would indicate no correction needed.

This study derives a relationship between filter mass loading and percent loss using results from previous studies of self-absorption and considers that the vast majority of the particulate loading rests on the top of the filter fiber media. Nevertheless, the mass loading itself may consist of particulate dust, radioactive particulates, and the filter material. The study by Higby (1984) calculated a minimum burial depth for an alpha particle to be lost due to absorption (100% loss) of about 3.7 mg/cm^2, mentioned previously, based on calculations for the range of 239-Pu alpha particles in glass fiber filters. Luetzelschwab et al. (2000) recommended assuming a 40% loss at a loading of 3.3 mg/cm^2 and a 28% loss for a loading of 2.3 mg/cm^2. The mass loadings in the Luetzelschwab et al. (2000) study included dust loading plus the mass of the filter front layer and are somewhat less when the filter front layer is eliminated. These data sets were previously studied and reported by Smith et al. (2011). This report describes our investigations of data presented in Higby (1984), Luetzelschwab et al. (2000), Huang et al. (2002), Barnett et al. (2009b), Smith et al. (2011), and Hogue et al. (2018). A summary of the mass loading aspect is provided in Table 1.

Table 1. Summary of Mass Loading Reports from All Sources

Summary of Mass Loading	Source
Glass fiber filters, aerosol generated particles from stock particle suspensions, counting losses due to burial in the filter matrix, filter matrix excluded.	Higby 1984
Bi-layer fiberglass filters, particles collected from air, counting efficiencies reported as a result of absorber thickness. The areal density of the front layer of the filter is reported and can be separated from the reported results.	Luetzelschwab et al. 2000
Three types of membrane filters were used, particles suspended deposited by pneumatic dry dispersion. Thin layer deposits of radioactive aerosols were not significantly degraded by an underlying thick layer of dust. Filter matrix excluded.	Huang et al. 2002
Acrylic copolymer filters on a nylon substrate, particles collected from stack operations. The vast majority of particles remain on the top of the filter. Filter matrix excluded.	Barnett et al. 2009b
Acrylic copolymer filters on a nylon substrate, particles collected from stack exhaust streams. Examined light dust loading on filter material, filter matrix excluded.	Smith et al. 2011
Glass fiber filters, particles collected from occupational airborne radioactivity monitoring. Correction factors developed based on sampled activity to air volume followed by Monte Carlo modeling. Filter matrix excluded.	Hogue et al. 2018

2.0 Methods and Data

2.1 Research Studies Investigated

Because of the nature of radiation from alpha-emitting sources, a logical relationship between percent loss and mass loading would give loss as an exponential function of loading. At low levels of mass loading, there is a small percent loss. Once mass loading reaches a certain value, loss increases exponentially with increasing mass loading until loss is 100%.

In total, six research studies were combined and evaluated for their functions of mass loading on alpha particle losses. As expected, each study revealed an increase in percent loss of particles with increasing mass loading on the filter. Haung et al. (2002) and Luetzelschwab et al. (2000) reported 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 mg/cm^2 and up to 10 mg/cm^2). However, degradation in sample results including the front layer of the filter has been shown for sample loadings as little as ~0.4 mg/cm^2 and upwards of 40% self-absorption when the particulate matter is 3.3 mg/cm^2. Filter mass was excluded in the results developed herein.

Huang et al. (2002) reported a 100% loss of particles at 10 mg/cm^2, and additional self-absorption factors were calculated based on results published by Hogue et al. (2018) who reported results used to calculate losses at various levels. These two reports agree that 100% losses are in the 10 mg/cm^2 range.

2.2 Functions of Mass Loading

The available mass-loading functions reported are provided below. Barnett et al. (2009b) and Huang et al. (2002) are not included because they do not report any equations or functions.

2.2.1 Hogue et al. (2018) Assumptions

According to Hogue et al. (2018), to determine the correction factors used below in the equation for calculating self-absorption factors for varying mass loading quantities, the following principles apply:

1. At dust loading levels less than 0.1 mg/cm^2, the correction factor is 1.2.
2. At dust loading levels greater than 0.1 mg/cm^2 but less than 1.7 mg/cm^2, the correction factor is 1.4.
3. At dust loading levels between 1.7 mg/cm^2 to 9 mg/cm^2, the correction factor is 0.744 + 0.39255 × dust loading (mg/cm^2).

Using the derived correction factor guidelines identified above, the self-absorption factor (% losses of alpha particles) can be calculated as follows:

$$\text{Self-Absorption Factor} = 100 - [(1 \div \text{correction factor}) \times 100]\ (\%)$$

The self-absorption factor is subtracted from 100 (assuming 0% losses at or near 0 mg/cm^2 mass loading) to determine the percent loss of alpha particles for the given amount of loading (mg/cm^2).

A complete list of values calculated with this equation can be found in Table 2, Section 3.3.

2.2.2 Smith et al. (2011) Linear Equation

2.2.2.1 Linear Equation

Smith et al. (2011) derived a linear equation that used three data points: one from Higby (1984) and two from Luetzelschwab et al. (2000). The study by Luetzelschwab et al. (2000) gives the values of 28% loss at 2.3 mg/cm^2 and 40% loss at 3.3 mg/cm^2 when the front layer of the glass fibers is included in the loading, or 28% loss at 1 mg/cm^2 and 40% loss at 2 mg/cm^2 with dust loading alone. Higby's study gives the value of 100% loss at a thickness of 3.7 mg/cm^2. The equation for the linear graph is as follows:

$$y = 25.575x$$

where y is the percent loss of the particles, and x is the amount of mass loading. The coefficient of determination (R^2) for the linear relationship is 0.97. Using this linear model, Smith et al. (2011) found that EM samples had losses of less than 7%.

This linear graph is provided in Appendix A.

2.2.2.2 Exponential Equation

Smith et al. (2011) also derived an exponential equation that used the same three points mentioned previously. The equation for the exponential graph is as follows:

$$y = 16.551e^{0.4787x}$$

where y is the percent loss of the particles and x is the amount of mass loading. The equation fits the three data points with a R^2 of 0.99; it does not assume zero losses with no loading. Using this equation, the expected percent loss for PNNL filters with average loadings of 0.1 mg/cm^2 are 17% and loadings up to 0.24 mg/cm^2 are 19% (Smith et al. 2011). It was concluded that samples generally have losses of less than 19% using this conservative exponential model. This exponential graph also is provided in Appendix A.

2.2.3 Luetzelschwab et al. (2000) Data Points

Data included from Luetzelschwab et al. (2000) states that for a mass loading, which includes the areal density of the front layer of the filter, of 2.3 mg/cm^2, the calculated loss of alpha particles is 28%. Also included from this study is a reported 40% loss when a mass loading of 3.3 mg/cm^2 is present. When the front layer areal density of 1.3 mg/cm^2 is removed, then the losses are 28% and 40% for mass loadings of 2 mg/cm^2 and 1 mg/cm^2, respectively.

2.2.4 Higby (1984) Data Point

To reiterate, Higby's study in 1984 reported that "… the minimum burial depth for an alpha particle to be lost due to absorption is approximately 0.0037 gm/cm^2" (Higby 1984), which means if graphed, there is 100% loss at 3.7 mg/cm^2. It also was reported that "… a correction which assumes 10–15% losses would ensure that concentrations of airborne alpha emitting radionuclides would not be underestimated by collection and analysis on glass-fiber filters." This is the bases for the 0.85 correction currently used by EM. The Higby study did not consider additional dust loading from non-radioactive material as noted in Luetzelschwab et al. (2000).

2.3 Compiling the Data

By using the respective equations and data sets, a master list of % Loss in relation to total Mass Loading was compiled and labelled with each credited researcher. Table 2 represents the set of data points investigated in this study.

Table 2. Complete List of Data Points from All Sources

Mass Loading (mg/cm^2)	Reported % Loss	Source
10	100	Huang et al. 2002
10	100	Hogue et al. 2018
9	79	Hogue et al. 2018
8.5	75.5	Hogue et al. 2018
8	74.3	Hogue et al. 2018
7.5	72.9	Hogue et al. 2018
7	71.4	Hogue et al. 2018
6.5	69.7	Hogue et al. 2018
6	67.7	Hogue et al. 2018
5.5	65.5	Hogue et al. 2018
5	63	Hogue et al. 2018
4.5	60.2	Hogue et al. 2018
4	56.8	Hogue et al. 2018
3.7	100	Higby 1984
3.7	54.4	Hogue et al. 2018
3.5	52.8	Hogue et al. 2018
3	48	Hogue et al. 2018
2.5	36	Hogue et al. 2018
2	29	Hogue et al. 2018
2	40	Luetzelschwab et al. 2000
1	28	Luetzelschwab et al. 2000
0.24	6.1	Smith et al. 2011
0.2	0.001[a]	Barnett et al. 2009b
0.1	0.001[a]	Huang et al. 2002
0.1	5	Huang et al. 2002
0.1	24	Hogue et al. 2018
0.05	17	Hogue et al. 2018
0.09	2.3	Smith et al. 2011
1.30E-12	0.001[a]	Smith et al. 2011

[a] Near-zero value to represent no observed self-absorption as reported by the source reference.

The data provided in Table 2 then were graphed multiple times using various mathematical functions. The resulting graphs are provided in Appendix B.

3.0 Results

3.1 Trinomial Functions

By calculating all percent losses of alpha particles due to mass loading, the following equation was derived from the trinomial function that was created when all data points were plotted on the same graph.

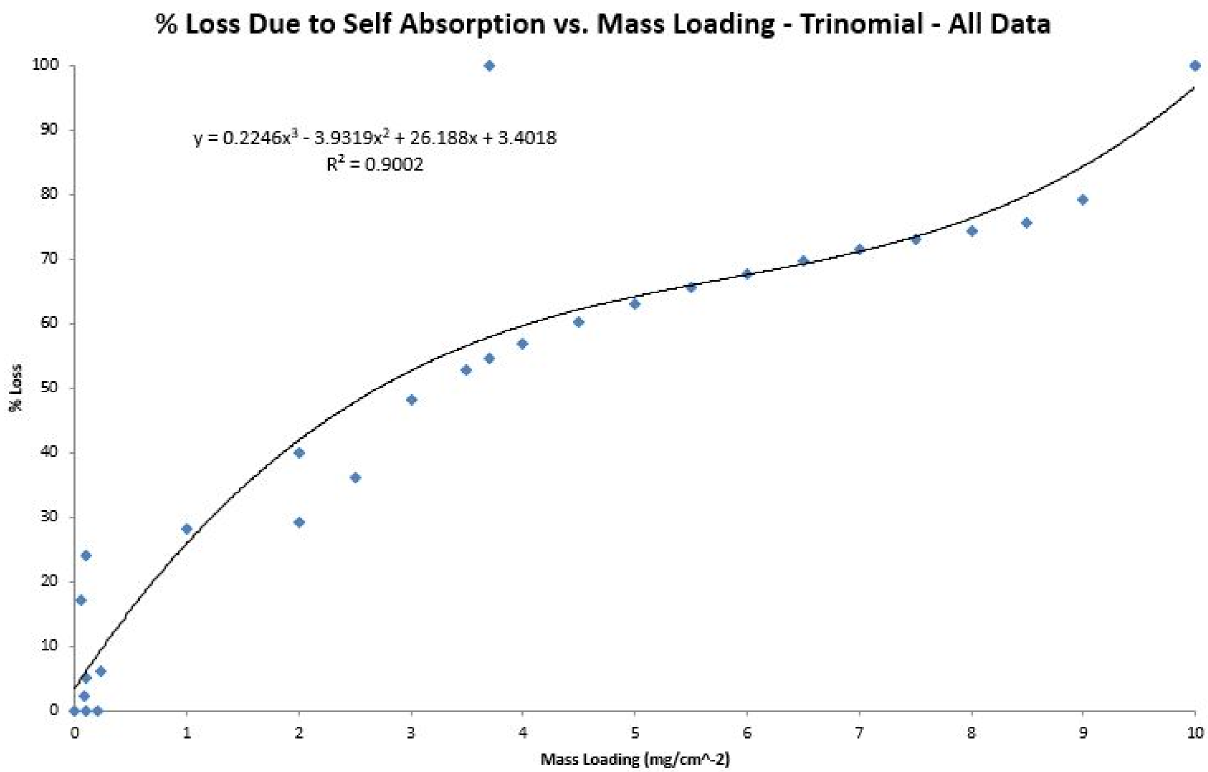

Figure 1. Trinomial Relationship of All Data Sources; Data Found in Table 2

The equation of the Figure 1 graph is as follows:

$$y = 0.2246x^3 - 3.9319x^2 + 26.188x + 3.4018$$

where the y-axis is percent loss and the x-axis is the mass loading (mg/cm^2). The value of 3.4018 at the end of the formula represents the total percent loss of ~3.4% at (or near) 0 mg/cm^2 mass loading.

This is significant because under the 0.85 correction factor currently used, it is estimated that at or near 0 mg/cm^2 mass loading, there is still 10–15% losses in alpha particles (Higby 1984). While this relationship supports that correction factor, it is overtly conservative and overestimates loss.

Also created was a forced zero graph that assumes there is 0% losses at 0 mg/cm^2 mass loading, as shown in Figure 2.

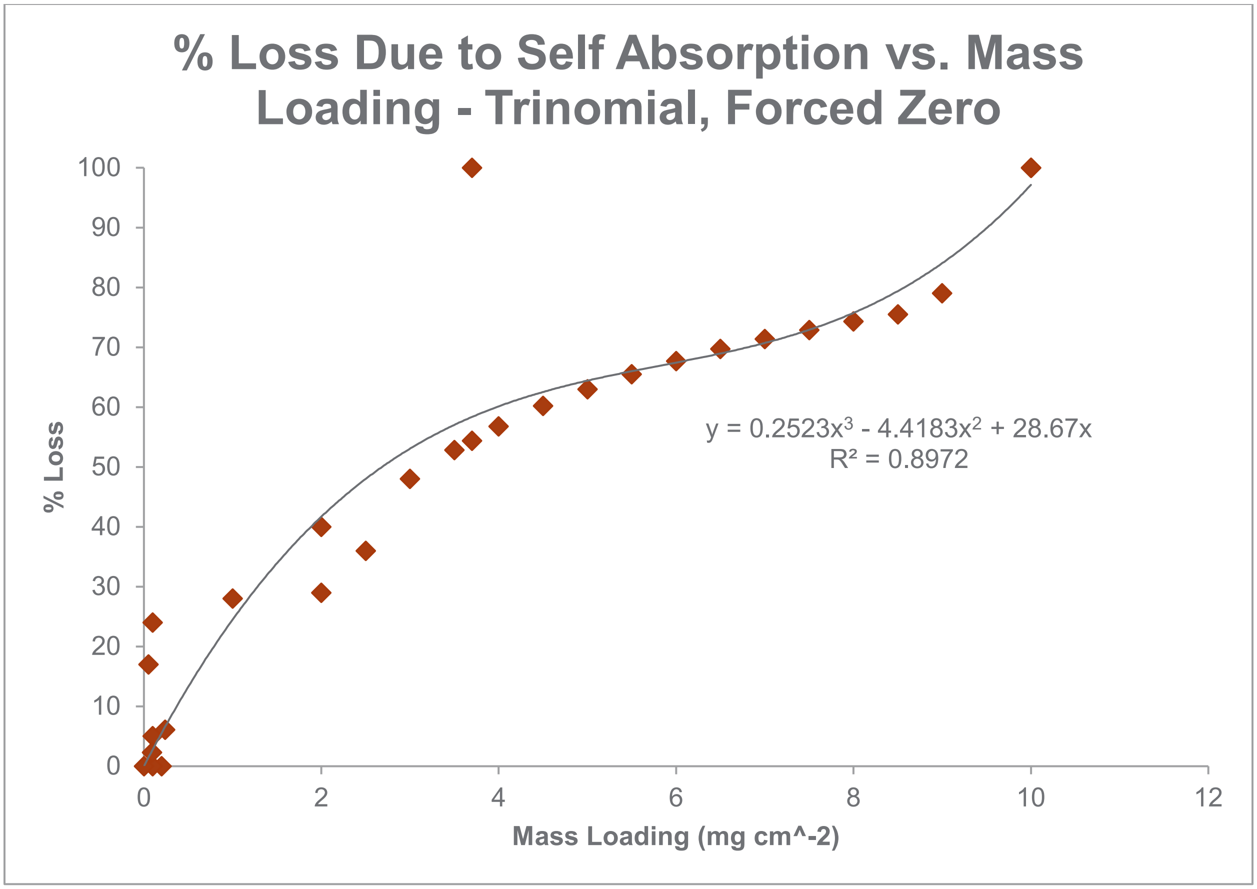

Figure 2. Trinomial Relationship of All Research Data Forced to Zero Intercept

The equation of the trinomial relationship found above is as follows:

$$y = 0.2523x^3 – 4.4183x^2 + 28.67x$$

3.2 Data Without Higby

Due to the results reported by the other researchers cited in this report, it is no longer surmised that mass loading has 100% loss effects at 3.7 mg/cm^2. Hence, if the Higby data point is removed, it being considered as a conservative outlier, and the relationship changes as shown in Figure 3 and discussed below (and the R^2 improves).

In this graphed trinomial relationship, the trendline represents the data well with an improved R^2 value of 0.97, and a percent loss of alpha particles of 4.4% when there is (or near) 0 mg/cm^2 mass loading.

A forced zero intercept graph excluding the Higby point is provided in Appendix B.

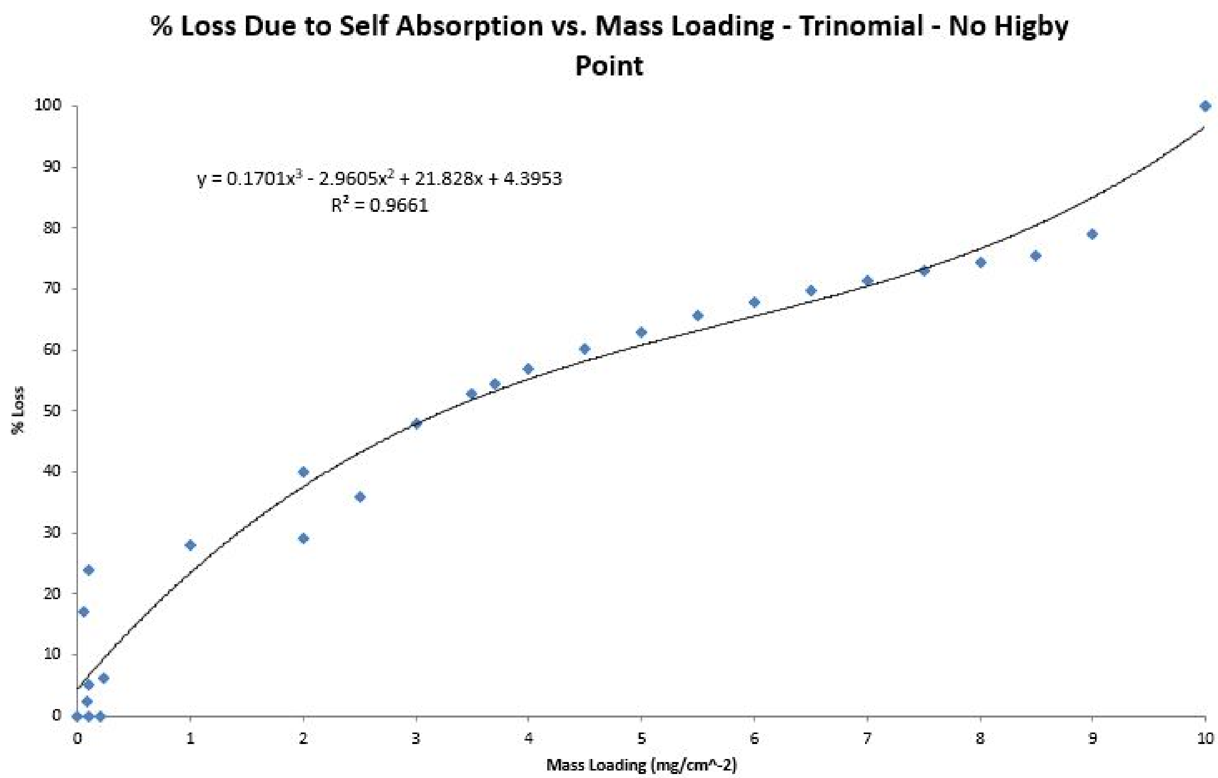

Figure 3. Trinomial Relationship of All Research Data, Without Higby Point

3.3 Data Derived from Trinomial Relationships

The trinomial relationships produced two functions that serve to calculate percent loss in a forced zero and near zero environment for mass loading on the particulate filters. Table 3 and Table 4 present data derived when the data from Table 2—all research data compiled—are inserted into the functions from Figures 1 and 2, as well as a calculated difference of what the functions produce, versus data that were reported by the six research studies. The data are in the same order as compiled in Table 2 but without the credited researchers list.

By observing Table 3 (where all the data are used, non-forced zero function), the following reported mass loading values of 3.7, 2.5, 2, 0.24, 0.2, 0.1, 0.1, 0.05, 0.09, and 1.30E-12 mg/cm^2 differed the most (greater than 25% difference) from the function provided by the trinomial relationship.

Table 3. Non-Forced Zero Intercept Results Calculated with Trinomial Function Using All Data

Non-Forced Zero Intercept: $Y = 0.2246x^3 - 3.9319x^2 + 26.188x + 3.4018$			
Loading (mg/cm^2) (x)	**Research Study Results (Y, % Loss)**	**Results from Calculated Equation(Y, %Loss)**	**% Difference from Reported Research Values**
10	100	96.7	-3.3%
10	100	96.7	-3.3%
9	79	84.3	6.8%
8.5	75.5	79.9	5.8%
8	74.3	76.3	2.6%
7.5	72.9	73.4	0.7%
7	71.4	71.1	-0.4%
6.5	69.7	69.2	-0.7%
6	67.7	67.5	-0.3%
5.5	65.5	65.9	0.6%
5	63	64.1	1.8%
4.5	60.2	62.1	3.1%
4	56.8	59.6	5.0%
3.7	100	57.8	-42.2%
3.7	54.4	57.8	6.3%
3.5	52.8	56.5	7.1%
3	48	52.6	9.7%
2.5	36	47.8	32.8%
2	29	41.8	44.3%
2	40	41.8	4.6%
1	28	25.9	-7.6%
0.24	6.1	9.5	55.1%
0.2	0.001	8.5	848292.1%[a]
0.1	0.001	6.0	598050.6%[a]
0.1	5	6.0	19.6%
0.1	24	6.0	-75.1%
0.05	17	4.7	-72.3%
0.09	2.3	5.7	149.0%
1.30E-12	0.001	3.4	340080.0%[a]

[a] Result not meaningful since the reported loss is essentially zero.

Table 4. Forced Zero Intercept Results Calculated with Trinomial Function

Forced Zero Intercept: $Y = 0.2523x^3 - 4.4183x^2 + 28.67x$			
Loading (mg/cm^2) (x)	Research Study Results (Y, % Loss)	Results from Calculated Equation (Y, % Loss)	% Difference from Reported Research Values
10	100	97.27	-2.8%
10	100	97.27	-2.8%
9	79	84.1	6.4%
8.5	75.5	79.4	5.2%
8	74.3	75.8	2.0%
7.5	72.9	72.9	0.0%
7	71.4	70.7	-0.9%
6.5	69.7	69.0	-1.0%
6	67.7	67.5	-0.4%
5.5	65.5	66.0	0.8%
5	63	64.4	2.3%
4.5	60.2	62.5	3.9%
4	56.8	60.1	5.9%
3.7	100	58.4	-41.6%
3.7	54.4	58.4	7.3%
3.5	52.8	57.0	8.0%
3	48	53.1	10.5%
2.5	36	48.0	33.3%
2	29	41.7	43.7%
2	40	41.7	4.2%
1	28	24.5	-12.5%
0.24	6.1	6.6	8.7%
0.2	0.001	5.6	555828.6%[a]
0.1	0.001	2.8	282206.9%[a]
0.1	5	2.8	-43.5%
0.1	24	2.8	-88.2%
0.05	17	1.4	-91.6%
0.09	2.3	2.5	10.6%
1.30E-12	0.001	3.7E-11	-100.0%[a]

[a] Result not meaningful since the reported loss is essentially zero.

By observing Table 4 in which all the data are used (i.e., the forced zero function), the following reported mass loading values of 3.7, 2.5, 2, 0.2, 0.1, 0.1, 0.1, 0.05, 1.30E-12 mg/cm^2 differed the most (greater than 25% difference) from the function provided by the trinomial relationship.

Using all the data, both the non-forced zero intercept function and forced zero intercept function had similar mass loading values that differed in calculated losses (greater than 25% different) the most from values in Table 2. Those compiled values are shown in Table 5. It appears that as the mass loading values get smaller, the differences from the trinomial function are the greatest. It suggests that at low mass loading values, variations in actual losses may be difficult to ascertain even though they are expected to be small.

Table 5. Mass Loading Values Calculated by Graph Functions that Differed More than ± 25% from Reported Research Values

Loading (mg/cm^2)	Average % Difference of Non-Forced and Forced Zero Trinomial Functions	Source
3.7	-41.9%	Higby 1984
2.5	33.1%	Hogue et al. 2018
2	44.0%	Hogue et al. 2018
0.2	702060.4%[a]	Barnett et al. 2009b
0.1	440128.7%[a]	Huang et al. 2002
0.1	-81.7%	Hogue et al. 2018
0.05	-82.0%	Hogue et al. 2018

[a] Result not meaningful because the reported loss is essentially zero.

3.4 Other Functions Used to Evaluate Source Data

The focus of Section 3 thus far has been the trinomial function. As stated previously, the same data source set was used to generate additional graphs evaluating linear, polynomial, and exponential relationships (see Appendix B).

The functions shown in Table 6 were derived from the graphs using a compilation of all research data (see Appendix B).

Table 6. Functions Derived from Graphs of All Research Data Presented in Appendix B

Graph Type	Function Derived
Linear (Near Zero)	$Y = 8.7151x + 14.034$
Linear (Forced Zero Intercept)	$Y = 10.827x$
Exponential	$Y = 1.3568e^{0.587x}$
Polynomial (Near Zero)	$Y = -0.7311x^2 + 15.307x + 7.2379$
Polynomial (Forced Zero Intercept)	$Y = -0.958x^2 + 18.124x$

3.5 Comparison to Typical PNNL Mass Loading

As reported in Smith et al. (2011), the typical mass loading from stack emissions at PNNL facilities is 0.09 mg/cm^2. Table 7 presents losses from the four trinomial equations for the PNNL nominal mass loading range as well as those mass loadings that would result in 15% losses (i.e., a correction factor of 0.85). Using the trinomial equation for all data and non-forced and forced zero results, the losses are calculated to be 5.7% and 2.5%, respectively. Similarly, for the trinomial equation without the Higby data, the non-forced and forced zero results show the calculated losses to be 6.3% and 2.2%, respectively. At the 0.09 mg/cm^2 mass loading, the non-forced zero functions result in losses just slightly greater than the 5% value in ANSI/HPS N13.1-2011, while the forced zero function results are clearly less than the 5% value.

The self-absorption correction factor resulting from mass loading on a filter is just one of several correction factors used at PNNL. Appendix C provides additional information and detail when all of the applicable corrections factors are used; and it shows the impacts and implications of varying just the self-absorption correction factor from 0.85 to 0.95 and to 1.

Table 7. Comparison of Various Mass Loads Versus Calculated Percent Loss

Mass Loading (mg/cm^2)	Trinomial All Data (% Loss)	Trinomial All Data – Forced Zero (% Loss)	Trinomial Minus Higby Data (% Loss)	Trinomial Minus Higby Data – Forced Zero (% Loss)
0	3.4	0.0	4.4	0.0
0.09	5.7	2.5	6.3	2.2
0.21	8.7	5.8	8.9	5.1
0.24	9.5	6.6	9.5	5.8
0.48	**15.0**	12.7	14.1	11.2
0.52	16.0	13.8	**15.0**	12.1
0.57	17.1	**15.0**	15.9	13.2
0.66	19.0	17.0	17.5	**15.0**

4.0 Conclusions

This study presents a trinomial function that relates percent loss due to self-absorption to filter mass loading. Results documented in this report are based on data published by Higby (1984), Luetzelschwab et al. (2000), Huang et al. (2002), Barnett et al. (2009a, 2009b), Smith et al. (2011), and Hogue et al. (2018).

By using results from Hogue et al. (2018), additional self-absorption values were derived from the correction factor calculations reported. It was then possible to calculate multiple data points that fit the trinomial function properly along with the addition of data from the other researchers. This trendline is reported as follows, which assumes 3.4018% alpha particle losses at 0 mg/cm^2 mass loading:

$$Y = 0.2246x^3 - 3.9319x^2 + 26.188x + 3.4018$$

This equation provides a method that can be used to calculate the percent loss of alpha particles with evidence supported by using values from the six research studies mentioned previously.

Higby (1984) reported 100% losses at 3.7 mg/cm^2 mass loading. Use of this equation yields a calculated alpha particle loss of 57.8%.

Luetzelschwab et al. (2000) reported 28% losses at 1 mg/cm^2 mass loading, and 40% losses at 2 mg/cm^2 mass loading. The trinomial function instead calculated an alpha particle loss of 25.9% at 1 mg/cm^2, and 41.8% loss at 2 mg/cm^2; both results seem reasonable when compared to the reported values.

Under ANSI/HPS N13.1-2011, *Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stack and Ducts of Nuclear Facilities*, it is recommended and accepted 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 and reported. Results from this study, which incorporate data from six different studies, when graphed and curve fitted to the relationship of the data, suggest that the total percent loss per mass loading data point, and at 0 mg/cm^2, is in the 3.4%–4.4% range and is less than 5% with and without the Higby data point, respectively.

With the PNNL average sample mass of 0.09 mg/cm^2 and self-absorption losses in the 2.2%–6.3% range, the currently used EM correction factor of 0.85 is conservative and results in over-estimates of actual emissions. Although the ANSI/HPS N13.1-2011 guidelines state that self-absorption of less than 5% losses does not require a correction factor, the results show that in the non-forced zero cases, losses could be just greater than 5%. If we take this into account and correct all the data at 5%, the stack sample data is still conservative and corrected especially presuming the non-forced zero cases also are conservative. Therefore, based on the research results published since 1984, it is recommended that use of the 0.85 correction factor be discontinued and a correction factor of 0.95 should be used. This change still would provide conservative estimates of mass losses.

5.0 References

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Appendix A – Smith et al. Graphs

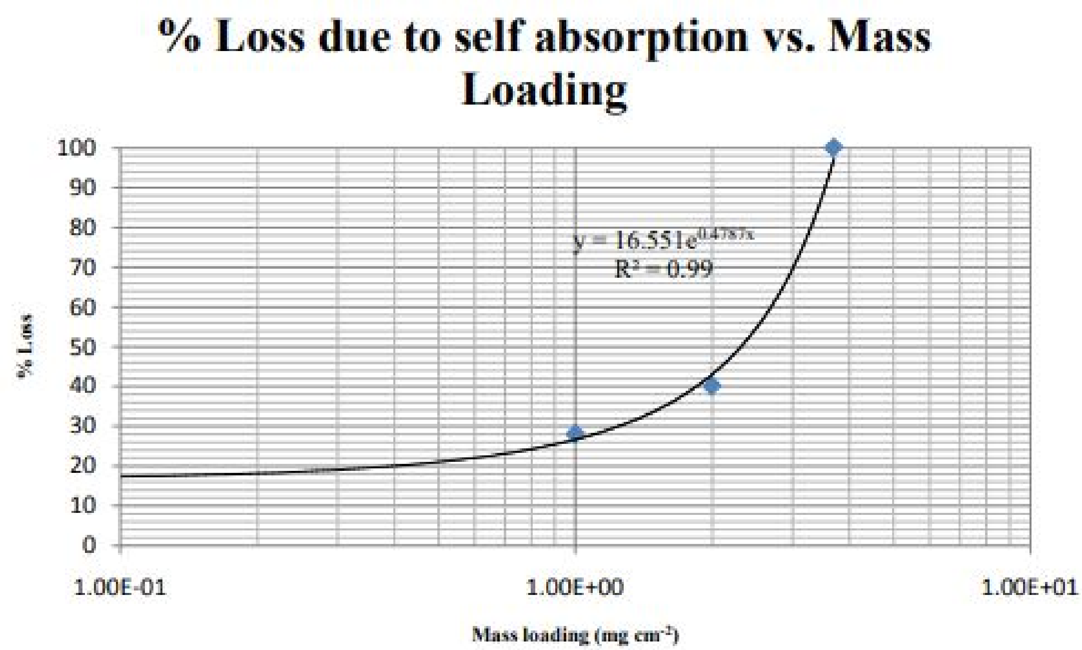

Figure A.1. Exponential Relationship of Percent Loss versus Mass Loading in Smith et al. (2011)

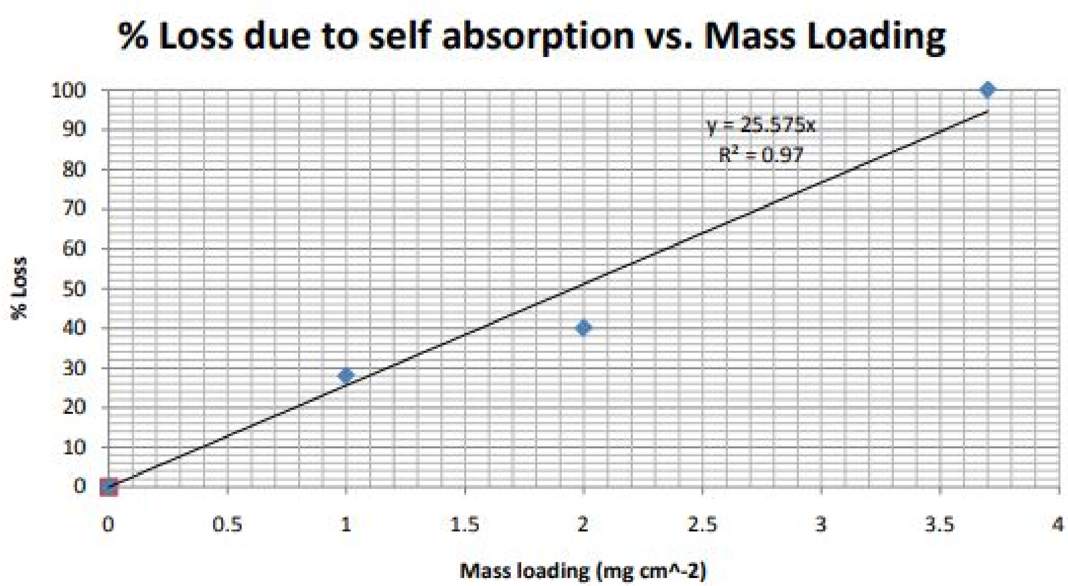

Figure A.2. Linear Relationship of Percent Loss versus Mass Loading in Smith et al. (2011)

Appendix B – Other Graphed Data Set Functions

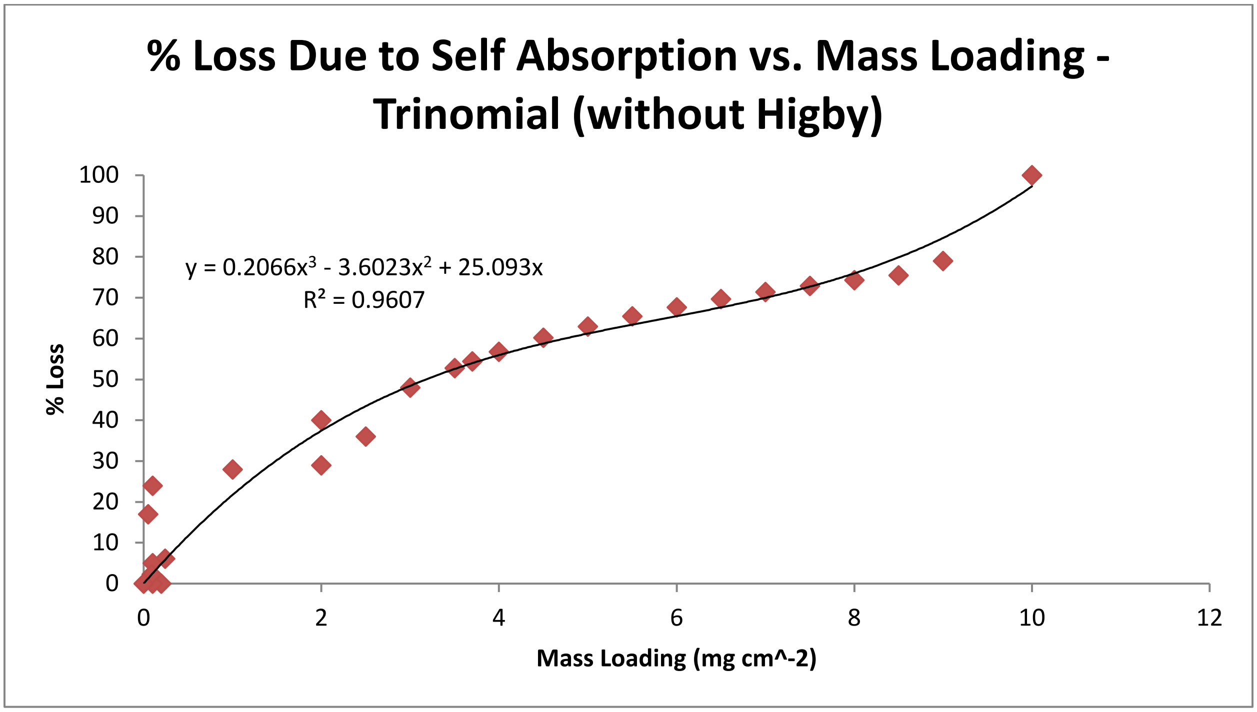

Figure B.1. Trinomial Relationship of All Data Forced to Zero Intercept, Without Higby Point

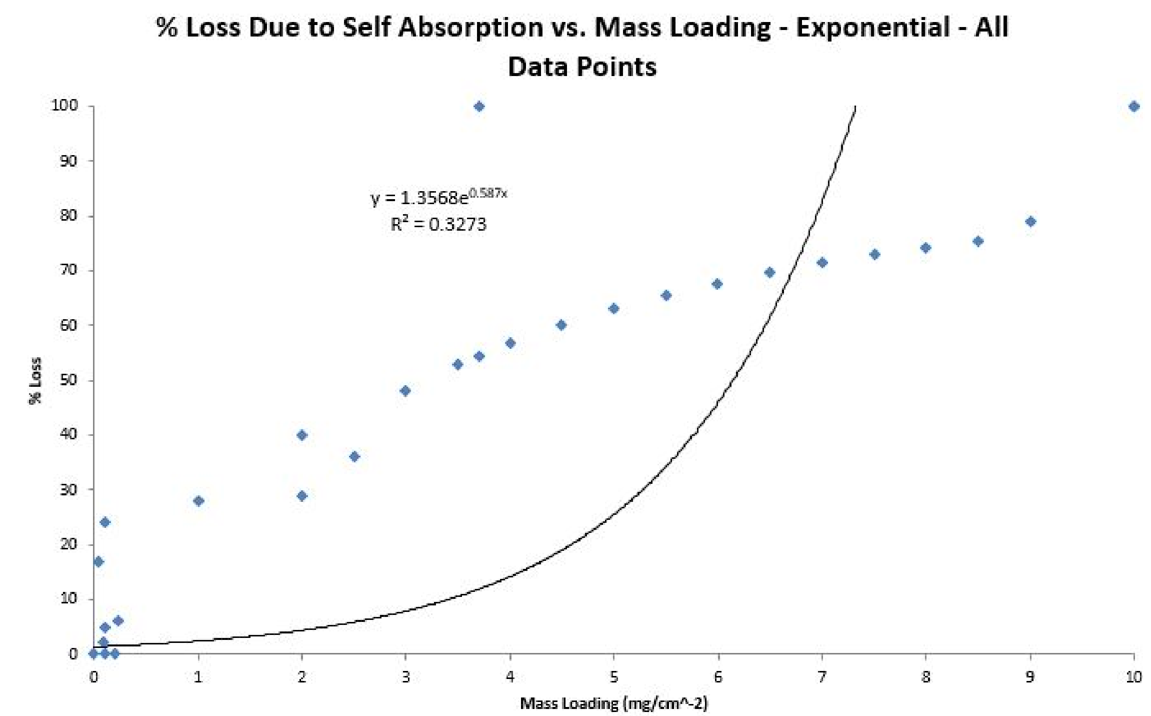

Figure B.2. Exponential Relationship of All Research Data

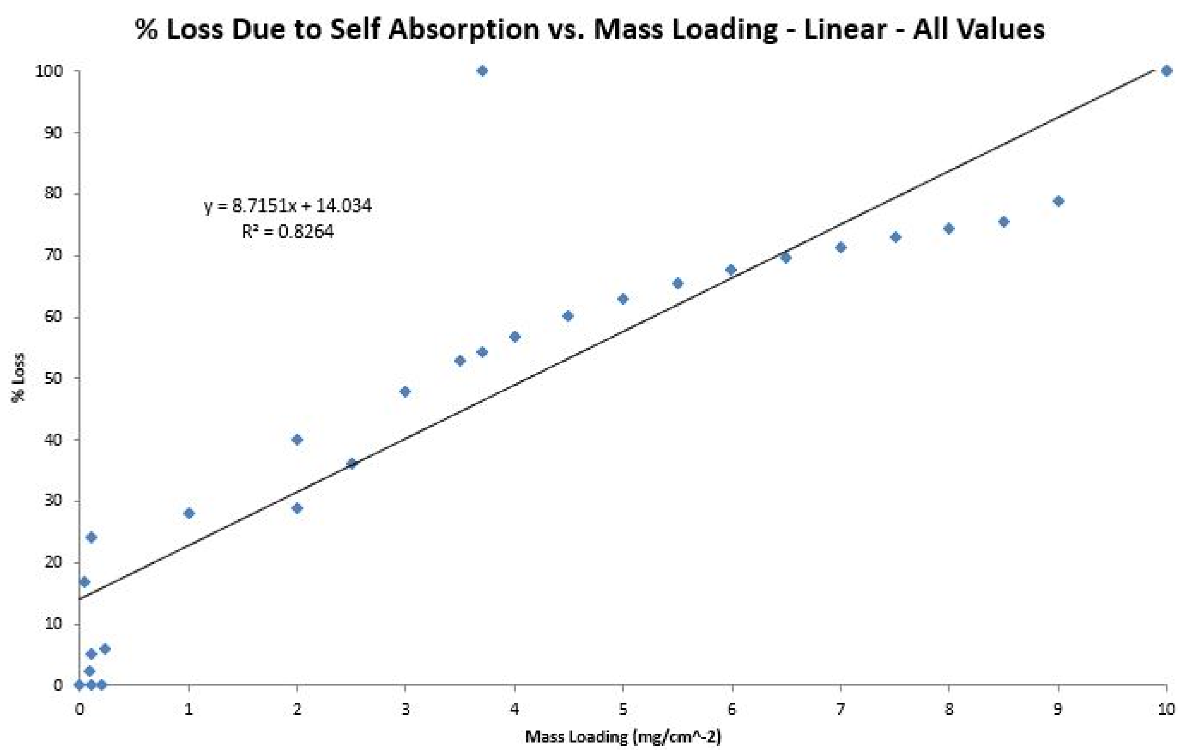

Figure B.3. Linear Relationship of All Research Data Near Zero

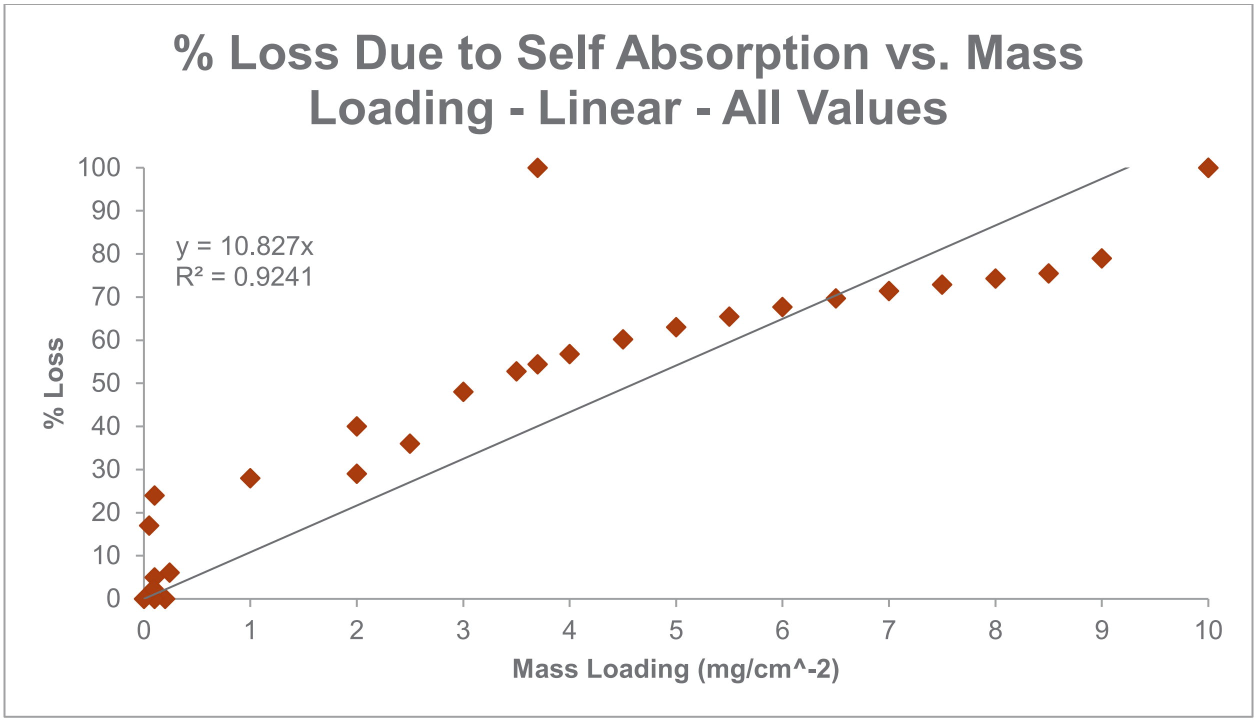

Figure B.4. Linear Relationship of All Research Data Forced to Zero Intercept

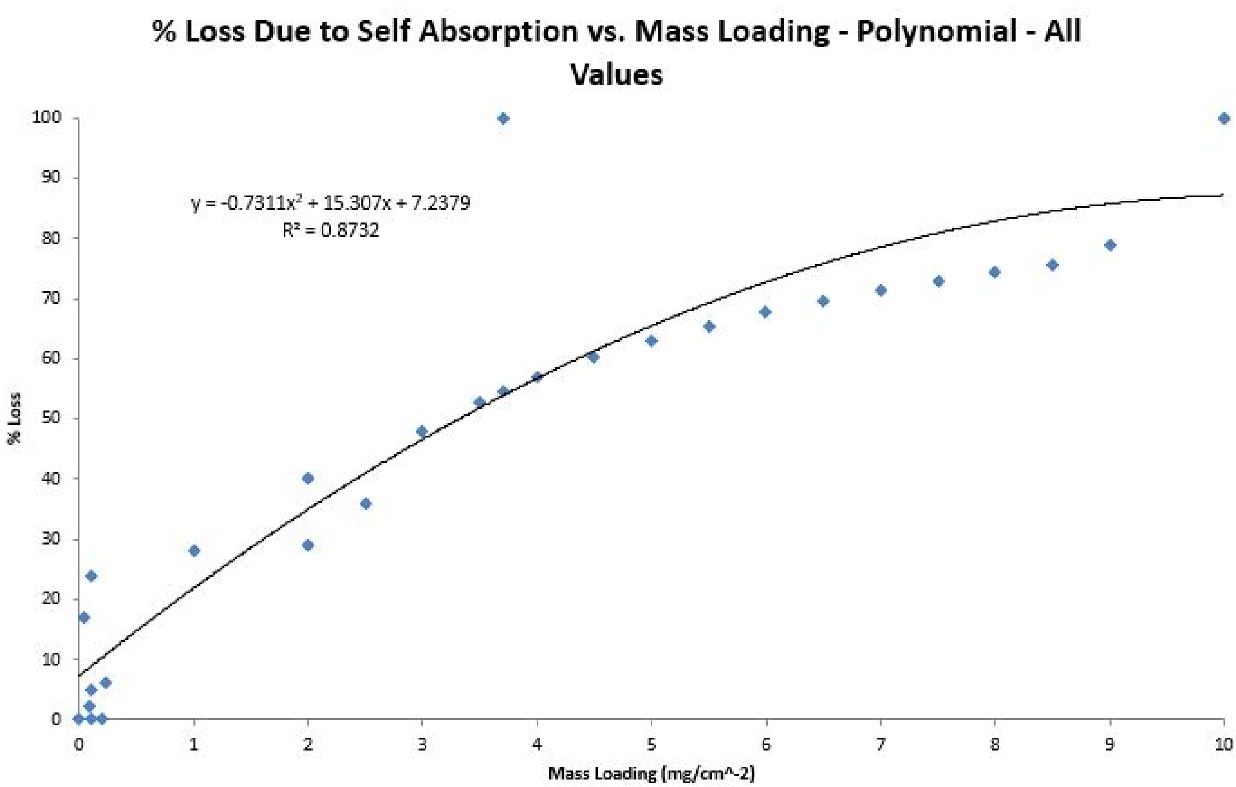

Figure B.5. Polynomial Relationship of All Research Data Near Zero

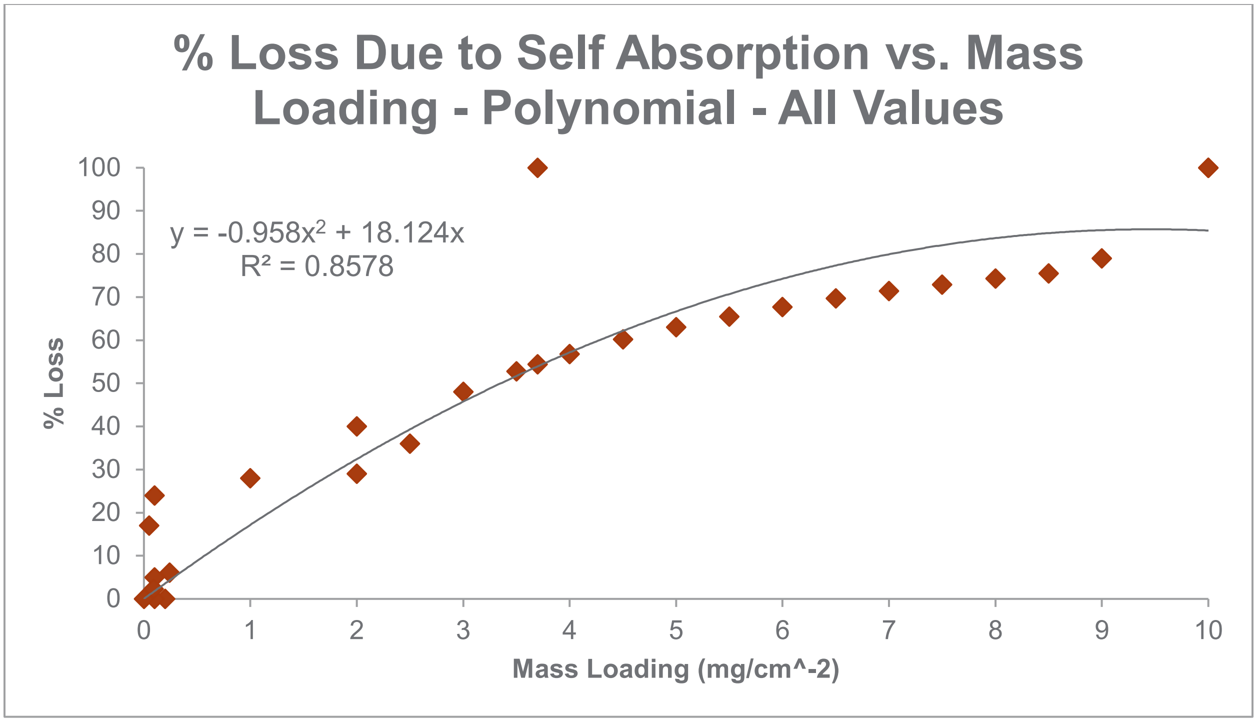

Figure B.6. Polynomial Relationship of All Research Data Forced to Zero Intercept

Appendix C – Correction Factor Discussion

Correction factors are commonly applied to prevent under-reporting of emissions and to maintain a conservative emission result (Barnett 2011). Types of correction factors that may be applied include:

1. Self-absorption
2. Transport efficiency
3. Sample collector media efficiency
4. Sampler operation efficiency
5. Exhaust traverse correction
6. Radioactive decay factor.

Correction factors typically are found in the denominator of the equation and are multiplicative in nature. The calculation and reporting of the final result should include the appropriate correction factors as discussed above. The total activity of a sample is then expressed as:

$$A_{Total} = A_{Sample}/\pi(E)_{(i)} \ (Bq)$$

where:

A_{Total} = total activity on sample in Becquerel

A_{Sample} = sample activity in Becquerel

$E_{(i)}$ = correction factors (i.e., those identified above).

Applying a self-absorption correction factor of 0.85, 0.95, and 1 with routine sample correction factors at Pacific Northwest National Laboratory shows that the total sample correction can improve from 28% to 14% and 9%, respectively, as shown in Table C.1. Similarly, when applying the self-absorption correction factors to results with an impacted (e.g., non-routine) sampler operation efficiency and exhaust traverse correction, the total sample correction can improve from 51% to 35% and 28% respectively, as shown in Table C.2.

Table C.1. Self-Absorption Correction Factor Comparison When Included With the Other Routine Operations Correction Factors

Self-Absorption	Transport	Media	Sample Operation	Traverse	Rad Decay	Total Correction Factor	Increase in Activity (%)
0.85	0.93	0.99	1	1	1	0.78	28%
0.95	0.93	0.99	1	1	1	0.88	14%
1	0.93	0.99	1	1	1	0.92	9%

Table C.2. Self-Absorption Correction Factor Comparison When Included With the Other Non-Routine Operations Correction Factors

Self-Absorption	Transport	Media	Sample Operation	Traverse	Rad Decay	Total Correction Factor	Increase in Activity (%)
0.85	0.93	0.99	0.94	0.90	1	0.66	51%
0.95	0.93	0.99	0.94	0.90	1	0.74	35%
1	0.93	0.99	0.94	0.90	1	0.78	28%

For routine operations, changing the self-absorption factor to 0.95 results in samples with about half of the activity as would typically be calculated. Not applying any self-absorption correction factor (i.e., 1.0) to routine operations results in samples with about two-thirds of the activity as would typically be calculated. Similarly, for non-routine operations samples, calculated activities would be approximately one-third to almost one-half of the sample activity result. Overall, the application of the various correction factors over a range of routine to non-routine situations shows the variability in reporting sampled emissions to the environment. A total correction factor range of 14–35% for a self-absorption factor of 0.95 is somewhat of an improvement over the 28–51% range for a self-absorption factor of 0.85, and some overlap still exists between the two. However, considering setting the self-absorption factor to 1, the total correction factor range of 9–28% provides essentially no overlap with the existing range.

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