# Appendix A

# **Air Deposition Modeling**

For purposes of this report, the U.S EPA's preferred model for regulatory applications AERMOD (v. 15181)<sup>1</sup> was used to estimate annual air deposition (g/m<sup>2</sup>/year) within 5 km of the facilities. AERMOD is a dispersion model which incorporates emissions and meteorological data for calculating deposition at receptor grid nodes. AERMOD has been demonstrated to be an effective tool for simulating air dispersion and deposition of PFOA and PFOS (Barton et al. 2010, Shin et al. 2011, Shin et al. 2012).

# 1.0 Model Setup

Data requirements for the AERMOD model include emissions characteristics of stack parameters (height, temperature, and flow rate), particle size information (for deposition calculations), receptor elevation data, and meteorological data. The input assumptions used for this modeling analysis are discussed in the following sections.

### 1.1 Model Domain and Spatial Resolution

For purposes of this report, the model domain for the air deposition model (Figure A1) was 13 kilometers x 14 kilometers centered between the Water Street and Northside Drive facilities. Air model receptors (specific locations where deposition estimates are calculated) were placed at 200-meter intervals across the domain with 20-meter receptor intervals adjacent to each facility for a total of 8,354 receptors. The receptors were placed in the modeling domain as to capture the maximum deposition impacts. The primary impacts and strongest gradients from particle deposition take place close to the facility and this domain is sufficiently large to capture these impacts.

### 1.2 Meteorological Data

For purposes of this report, a five-year meteorological data set (2006-2010) from the Bennington, Vermont airport (prepared by VTDEC) was used in this analysis. The meteorological data set consists of wind, temperature, precipitation, and cloud cover measurements collected from the airport meteorological station combined with upper air data (weather balloon soundings from the Albany, NY airport) for estimating atmospheric stability. More importantly, the AERMINUTE meteorological data processor for ASOS (Automated Surface Observation Stations) data was used in the 2006 – 2010 data processing to develop hourly average wind speed and directions to reduce the number of calms and missing winds in the meteorological data set.

<sup>&</sup>lt;sup>1</sup> A more recent version of AERMOD (v16216r) is available. The model version was not changed from the initial modeling because the changes in AERMOD from v15181 to v16216r primarily affected modeling if the ADJ\_U\* option in AERMET was used, which is not the case for this analysis.

One option that was considered for developing a modeling meteorological data set was to use meteorological data concurrent with facility operations (1969 – 2001). However, ASOS data collection methods suitable for use by AERMINUTE began in 2005 at the Bennington Morse State Airport and the meteorological measurement methods of the historical data are less sensitive than ASOS data collection of the 2006 – 2010 data set. The concurrent historical data set also had a significantly higher percentage of calm wind conditions (approximately 30% calm winds as compared to 3.6% calms using AERMINUTE). Additionally, the nearest airport with a concurrent data set suitable for modeling is from Albany, NY. Although Albany is only approximately 30 miles from Bennington, due to differences in terrain and land use, the historical Albany meteorological data set was not sufficiently representative of Bennington wind patterns for use in this analysis.

The USEPA<sup>2</sup> recommends the use of a five-year meteorological data set to adequately capture interannual meteorological variability provided that the data are reasonably representative for the analysis. Given that the objective of the analysis is to estimate long term deposition around the facilities, the representativeness and robustness of the meteorological data set are a key variable. The use of this AERMINUTE meteorological data set reduced the number of calm hours and therefore increased the number of model calculation hours. As such, given that the data set was demonstrated to be representative, its use is more appropriate than to use concurrent meteorological data.

The 2006-2010 wind rose for the modeling data set is shown in Figure A2. The data set shows a predominant westerly component and well as winds from the south and northwest consistent with the climatology of the northeastern United States<sup>3</sup>. Figure A3 shows the wind rose for 2000 – 2012 which is the period of record for the Morse Airport. Comparing Figures A2 and A3 demonstrates that the modeling data set is representative and also shows the substantially reduced number of calm hours in the modeling data set through the use of AERMINUTE.

### 1.3 Stack Data

The stack parameters required for the modeling analysis include emission rate, stack height, stack temperature, stack gas velocity and stack diameter for each emission source in the analysis.

### 1.3.1 Air Emissions

Air emissions of PFOA occur during the baking process within the product manufacturing. Attachment A-1 "PFOA Emissions from the Glass Fabric Coating Process" provides a detailed explanation of the coating process and the mechanisms affecting PFOA emissions.

<sup>&</sup>lt;sup>2</sup> Appendix W of 40 CFR Part 51 – Section 8.3.1.1

<sup>&</sup>lt;sup>3</sup> Climate of Vermont. National Climatic Data Center,

www.ncdc.noaa.gov/climatenormals/clim60/states/Clim\_VT\_01.pdf

#### 1.3.1.1 Coating Process Summary

The Bennington, VT (Northside Drive) and North Bennington, VT (Water Street) facilities manufactured PTFE (polytetrafluoroethylene) coated glass fiber fabrics. The coated fabrics were produced in coating towers. The manufacturing steps associated with PFOA emissions from the coating towers include:

- a) Coating apply a water based PFTE coating (dispersion) to glass fiber fabric
- b) Drying heat the coated fabric to 200° to 300°F to remove water
- c) Baking heat the coated fabric to 550° to 650°F to destroy or remove surfactants
- d) Fusing heat the coated fabric to 600° to 700°F to cure the PTFE and bond it to the glass fabric

The water-based PTFE coatings are called dispersions. The PTFE in the dispersions consists of small granules approximately 0.25 microns in diameter. Surfactants are used in dispersion formulations to keep the PTFE granules suspended in water while the coating is applied to the glass fabric. APFO (ammonium perfluorooctanoate) is one of surfactants used in the dispersion formulations. Most of the APFO is destroyed in the baking step where the coated fabric is heated to destroy surfactants used in the dispersions. However, a small fraction of the APFO is released in the coating towers at the Water Street plant were equipped catalytic oxidizers (abators) which reduced the amount of PFOA released to the atmosphere.

#### 1.3.1.2 **PFOA Air Emission Calculations**

For purposes of this report, PFOA air emissions are assumed to be a function of:

- 1. The amount of dispersions used to coat the glass fabric;
- 2. The amount of APFO used in the dispersion formulations;
- 3. The fraction of APFO released as PFOA from the coating towers;
- 4. Abator PFOA control efficiency (CE).

The data sources for estimating PFOA emissions for purposes of this report are:

- 1. Company records of annual dispersions use;
- 2. The average APFO content for high-PFOA content dispersions calculated from the Merrimack, NH facility data<sup>4</sup>;
- 3. Air partition factors based on industry trade group testing (Society of Plastics Industry, 2005 DPMB Report) that occurred at the SGPP Merrimack, NH plant.
- 4. Stack testing on the North Bennington facility coating Tower R.

Note: the Northside Drive facility did not have abator controls.

Air emissions for this report are calculated as:

<sup>&</sup>lt;sup>4</sup> Based on PFC usage data submitted by Saint Gobain Performance Plastics to the New Hampshire Department of Environmental Services in a letter to Clark Freise, Assistant Commissioner, dated July 22, 2016 as confidential business information.

PFOA <sub>air</sub> = dispersions (lbs) \* PFOA content (ppm) \* air partition factor (%) \* (1 – abator CE) (%)

The Merrimack, NH facility dispersions data from 2004 – 2005 estimated average APFO content was 2,000 ppm (wet basis). This is a conservative estimate (e.g., unlikely to underestimate) of APFO content for the Bennington and North Bennington facilities.<sup>5 6 7 8</sup>

For purposes of this report, the APFO air partition factors from the DPMB reports were used in the Bennington and North Bennington PFOA emissions calculations. The APFO partition factors measured were 8.8% for 1 meter towers and 11% for 2 – 4 meter towers. A detailed discussion of the testing results from the DPMB study and their application to this analysis are provided in Attachment A-1.

The relocation of operations from the Northside Drive facility to the Water Street facility also included adding pollution control equipment (abators) at the Water Street facility. The abators are thermo-catalytic control devices applied to the exhaust gases from the stacks. The 46% PFOA destruction rate for the abators was determined based on particulate testing of the R coating tower (TRC Environmental Corporation, 1999) with and without the abator in service and stack testing at the Merrimack, NH plant which showed the particulate control is indicative of PFOA control. A detailed assessment of abator control is provided in Attachment A-1.

Table A-1 shows the annual modeled PFOA emissions for the Northside Drive and Water Street facilities. Note that ChemFab moved from Northside Drive to Water Street in June 1978. For purposes of this report, all of the 1978 emissions were assumed to occur at the Water Street facility, instead of modeling partial year operations at each facility.

Model sensitivity: model estimated deposition is linear with respect to emission rate, consequently the modeled emission rate is an important parameter for deposition estimates.

<sup>&</sup>lt;sup>5</sup> SGPP accounted for its dispersions usage on a dry weight basis (solids only). APFO concentrations are reported on a wet basis. Dispersions typically contain 60% PTFE solids by weight on a wet basis with the balance (40%) of the dispersion formulation consisting of water and surfactants. In the calculations for APFO emissions, the SGPP dry weight dispersion use rates were divided by 0.6 to convert to dispersion use rates from a dry basis to a wet basis to be consistent with the APFO concentration data – 2,000 ppm (wet) / 0.6 = 3,000 ppm (dry).

<sup>&</sup>lt;sup>6</sup> Company records show that many of the dispersions used at Merrimack, NH were also used at the Bennington and North Bennington facilities.

<sup>&</sup>lt;sup>7</sup> Industry references (Drobny, 2001 and Ebnesajjad, 2000) suggest the typical APFO content in the dispersions is 0.1% (1000 ppm), and <2000 ppm, respectively.

<sup>&</sup>lt;sup>8</sup> As part of the 2010/2015 US EPA PFOA Stewardship program, six dispersion manufacturers worked with US EPA to reduce PFOA emissions by 95% from baseline emissions. APFO concentrations reported to US EPA (US EPA PFOA Stewardship Program, 2006) in the baseline emission report for dispersion manufacturers used by SGPP were 0.1364%, 0.0420% and 0.097% for Asahi (Fluon), Diakin and DuPont respectively for the 2000 to 2004 baseline period.

#### 1.3.2 Stack Parameters

The model uses stack parameter data (height, flow rate, diameter, and temperature) in its dispersion calculations. Stack location, height, flow rate, and diameter data were developed from facility data and from the facility permits and correspondence posted on the VTDEC website (ChemFab-Files-DEC-AIR .zip). The Water Street facility made several process modifications during its years of operation including adding and removing processing lines, relocating stacks, and modifying stack parameters. Stacks/ process lines were identified by letter, and the final permitted stack/ tower was Tower S. Some process lines shared emission points (e.g., lines B, C, and D). The Northside Drive facility assumed a single stack assumed to be the same as Stack A at the Water Street facility, as no site-specific stack parameters were available for the Northside Drive facility. Table A-2 lists the stack parameters used in the modeling for the Water Street facility.

For purposes of this report, the air emissions from the Water Street facility were assumed to be emitted primarily from the process stacks. However, based on facility inspections by VTDEC personnel, visible emissions were seen from the cupola vents, which could potentially have PFOA emissions. To account for this possibility, 5% of total emissions were assumed to be uncontrolled (i.e., not through the abators, modeled as a fugitive source) at the Water Street facility. Because very limited data exist for the Northside Drive facility and because production was much less at Northside Drive as compared to Water Street, for purposes of this report, it was assumed that all of the emissions at the Northside Drive facility were emitted from a single stack.

For purposes of this report, facility drawings from various times were used to group together similar stacks (both in location and stack parameters). Annual emissions from 1978 – 2001 were modeled from the stacks which were in operation during each year. Annual emissions were apportioned to the operating stacks based on abator capacity (mmbtu/hr). The stack parameters from 1998 (the year with the highest estimated PFOA emissions) were assumed for all years, instead of trying to model each years' configuration separately. Figure A4 shows the modeled air emissions sources for the Water Street facility.

For purposes of this report, stack temperatures were assumed to be 575 K (575 F) for stack gas exiting the abators. For the dilution stacks (P, G, and RS), the stack temperature was assumed to be 300 K (80 F) and the stack flow rates account for the dilution air. For 1998 emissions, the abator control efficiency was not applied to Towers P & G, as the abators were bypassed for those towers in 1998.

Model sensitivity: for annual average deposition, the model is not very sensitive to small changes in stack parameters. Consequently, the simplifying assumptions regarding facility stack layout over time are reasonable for this analysis.

#### 1.3.3 Building Downwash

Building downwash occurs when the stack emissions get entrained in the turbulence wake zones in the vicinities of buildings. Building corner coordinates and heights are used to determine wind direction specific building downwash parameters. Building downwash effects increase ground-level concentrations

near the source. For purposes of this report, the 1998 stack / building configuration (Figure A4) was used to develop the building downwash parameters used as an input to the AERMOD model.

## 1.4 Receptor Data

In addition to x, y coordinates for each receptor, receptor terrain elevations are incorporated in the model for purposes of this report. National Elevation Dataset (NED) 1/3 arc-second (10-meter resolution) terrain data available from the USGS National Map were processed using the AERMOD terrain preprocessor AERMAP (v. 11103).

Model sensitivity: while including terrain in the modeling analysis is important, changes in elevation either from an updated terrain data base or due to land use changes will have a minimal effect on modeled annual deposition. The groundwater modeling surface elevations used 30-meter resolution terrain data.

## 1.5 Particle Deposition Parameters

For estimating particulate deposition, the AERMOD model allows the user to input particle information in one of two ways. Method 1 is used when the particle size distribution is reasonably well known and a majority of the particulate are larger than 10 microns in diameter. Method 2 is used when the particle size distribution is not well known and the majority of the particulate is less than 10 microns in diameter.

For purposes of this report, Method 2 deposition was used for this modeling analysis. Perfluorooctanoate deposition parameters were taken from Barton et al., 2010, which assumed a fine particle fraction (particles < 2.5  $\mu$ m) of 0.61. These parameters represent the average measured particle size collected from a perfluorooctanoate monitoring program.

Model sensitivity: the modeled deposition results are very sensitive to the deposition parameters selected. Other data sources (Barton et al., 2006; Dreyer et al., 2015; Shin et al., 2012; Paustenbach et al., 2007) suggest a size distribution with a greater percentage of very small particles (< 1  $\mu$ m) than the one used in this analysis<sup>9</sup>. Modeled deposition decreases with a smaller particle size distribution, therefore this analysis has greater mass deposited on the grid than if deposition were represented with one of the other methods.

## 1.6 Model Output

The model was used to calculate annual deposition (total, wet, and dry) in g/m<sup>2</sup>/year. The 5-year average deposition at each receptor was multiplied by each year's annual PFOA emissions to calculate total deposition for input to the unsaturated zone model.

<sup>&</sup>lt;sup>9</sup> These other studies also used Method 1 deposition based on ambient particle size data. For approximately equivalent Method 1 and Method 2 deposition representation of these very small particle sizes, Method 2 results had greater deposition than Method 1. Because the particle size data were developed from ambient data (and not from stack testing), Method 2 was selected for this analysis.

# 2.0 Results and Discussion

The annual deposition for each year was used as input to the groundwater model (See Appendices B, C, and D for details). Air modeling files will be provided for VTDEC review.

## 2.1 Discussion

Running the air deposition model using reasonable estimates of annual PFOA emissions rates and representative meteorology provides a best estimate of PFOA air deposition given the data limitations for recreating past operations. The modeled annual deposition for years 1969 through 2001 are shown in Attachment A-2. Output for each year was used as input to the unsaturated zone model.

Dry deposition mechanisms account for virtually all (>99%) of the PFOA mass deposited by the model due to the inefficiency of washout (precipitation scavenging) on local-scale deposition.

## 2.2 Model Sensitivity

As discussed in Section 1, the modeled deposition is most sensitive to emission rates and deposition parameters. These parameters are felt to be reasonably representative with a bias towards overestimation (as compared to other candidate methods). As with any model, each model input value as well as the model formulation underscore the usefulness of the modeling analysis for understanding and explaining actual conditions. The goal of this modeling analysis was to develop reasonably likely estimates of PFOA deposition in the vicinity of the Bennington and North Bennington facilities using the best available data and accepted air modeling methods.

### 2.2.1 Model Selection

The AERMOD dispersion model is EPA's preferred model for regulatory applications. AERMOD is widely used and developing model input data (e.g., stack parameters, meteorological data, receptor data) is relatively straightforward as compared to other candidate models. AERMOD model performance is best for long term (annual) averaging periods.

### 2.2.1.1 AERMOD vs CALPUFF

For purposes of this report, a single meteorological year (2006) model sensitivity analysis was conducted to compare AERMOD results to CALPUFF results to address one of VTDEC's comments regarding model selection. CALPUFF's meteorological data requirements are more complex as compared to AERMOD's and in general, CALPUFF and AERMOD perform similarly for annual average concentrations as modeled results will follow annual wind patterns. However, due to the moderately complex local terrain there is a potential for annual deposition patterns to be different between the two methods.

Due to the limitations of the meteorological data used in CALPUFF (the CALPUFF meteorological data set did not use AERMINUTE, and consequently had 47% calms in the data set), the additional effort required to develop appropriate model inputs for CALPUFF appear to have limited benefit. A second concern is that CALPUFF treats deposition differently than AERMOD, so there are additional uncertainties with

respect to comparing CALPUFF and AERMOD results. AERMOD was the model selected for similar modeling analyses (Barton et al., 2006; Barton et al., 2010; Dreyer et al., 2015; Shin et al., 2012; Paustenbach et al., 2007).

#### 2.2.1.2 Meteorological Data

As discussed in Section 1.2, the 2006-2010 AERMET data set processed using AERMINUTE was considered to be the best choice for purposes of this report as compared to developing a concurrent meteorological data set from less representative sites (e.g., Albany, NY) or sites with high percentages of calm or missing data (Bennington sites pre-2005).

#### 2.2.1.3 Fogging / Condensation

VTDEC commented that observations of the emission plumes from the Water Street facility were frequently saturated (as noted by the water vapor in the plume) and that condensation and deposition may be an important fate and transport mechanism, especially during low wind conditions. The AERMOD model does not have the capability to explicitly model condensation, evaporation, or particle growth. However, more deposition occurs near the facilities for low wind conditions. Wet deposition is included in AERMOD, however wet deposition (through precipitation mechanisms) is a very small percentage of total deposition.

A fogging analysis was conducted using CALPUFF for two stacks at the facility Tower E (with abator) and Tower R (without abator). Fogging at Tower R was modeled to occur 20% of the 2006 meteorological data set. Fogging at Tower E was modeled at less than 5% of hours.

While the phenomenon of plume condensation may have been important for some stacks some percentage of time, overall, attempting to quantify and model fogging and condensation mechanisms is beyond the capability of the model physics.

### 2.2.2 Emission Estimates

The model-estimated deposition is proportional to the emission rates in the model. The modeling conducted for purposes of this report, incorporates annual variability in emissions based on annual dispersions usage, which is reasonable given that the dispersions are the source of the PFOA emissions. There is some uncertainty regarding the variability of PFOA content in dispersions as well as air partition factor and abator control efficiency. However, the values selected for these parameters for this analysis are reasonable based on the available data as discussed below.

The 2004 – 2005 Merrimack NH dispersions data set was considered the best available source of APFO content in dispersions formulation for purposes of this report. The estimated 2,000 ppm (wet) APFO content in the dispersions is a conservative estimate (e.g., unlikely to underestimate) of APFO content for the Bennington and North Bennington facilities. As noted above, the 2,000 ppm APFO concentrations is consistent with industry references and PFOA dispersion concentrations reported to EPA under the PFOA Stewardship program. In the DPMB report, the APFO dispersion used in a 1-meter tower the test had an APFO content of 1,130 ppm, and the 4-meter tower dispersion had an APFO content of 3,450 ppm. As

noted above, test conditions for the DPMB report were selected for measuring maximum actual emissions; so, it reasonable to expect that the APFO content of the dispersions used during testing would be slightly higher than average concentrations.

The abator control efficiency for PFOA emissions has some uncertainty as these control devices were not explicitly tested for PFOA control. However, as discussed in Section 1.3.1, the inferential evidence is compelling (notably PFOA's affinity to adhere to particulates and the effectiveness of particulate controls in controlling PFOA emissions, and that the abators controlled particulates) that PFOA emissions were reduced through the use of the abators.

Similarly, although there is some uncertainty in the air partition factors, these partition factors were developed for the exact processes occurring at the Bennington facilities, so the uncertainty around these air partition factors is low (i.e., there is high confidence in their representativeness).

### 2.2.3 Stack Parameterization

For purposes of this report, emissions were assumed to occur at a constant rate throughout the year. The Water Street facility generally operated 5 days per week. Because developing annual deposition estimates is the objective of the air modeling, the simplified representation of emissions (i.e., not incorporating daily or weekly variability) is not considered to contribute a significant amount of uncertainty to the analysis.

### 2.2.4 Deposition Parameters

The model estimated deposition (g/m2/yr) rates are very sensitive to the selected model representation of deposition. The AERMOD Method 2 deposition selection for this analysis resulted in more mass being deposited in the model domain than the other candidate methods.

## 2.3 Conclusions

The groundwater modeling results show reasonably good correlation with groundwater monitoring data. Consequently, because the PFOA mass deposited via the air modeling pathway provides input to the groundwater model, it may be concluded (albeit circumstantially) that the air deposition analysis reasonably recreates past PFOA air emissions and deposition.

# 3.0 References

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http://viewer.nationalmap.gov/basic/?basemap=b1&category=ned,nedsrc&title=3DEP%20View

### Table A-1

#### Modeled PFOA Annual Emission Rates Former ChemFab Facilities Bennington (Northside Drive) and North Bennington (Water Street), VT

		Annual	
		Dispersions	Annual PFOA
Facility	Year	Usage (lbs)	Emissions (lbs)
Northside Drive	1969	33,573	16
	1970	28,168	13
	1971	35,683	17
	1972	83,232	39
	1973	84,566	39
	1974	66,703	31
	1975	222,580	104
	1976	168,387	79
	1977	175,946	82
	Average Annual	Emissions	47
Water Street	1978	150,791	26
	1979	322,299	55
	1980	652,263	130
	1981	427,247	85
	1982	446,825	91
	1983	355,172	73
	1984	593,005	127
	1985	499,532	107
	1986	433,371	93
	1987	593,885	127
	1988	672,061	144
	1989	705,962	139
	1990	909,835	178
	1991	797,397	156
	1992	973,763	187
	1993	986,827	189
	1994	818,559	157
	1995	1,116,825	214
	1996	1,037,821	194
	1997	1,145,552	217
	1998	1,411,155	307
	1999	1,065,531	197
	2000	777,280	145
	2001	777,280	145
	Average Annual	Emissions	145

Notes:

2000 and 2001 estimated emissions are an average of 1978 - 1999 emissions. Usage data only available until May 2000.

# Table A-2Modeled Stack Parameters

North Bennington (Water Street), VT

		E	nglish units [1	L]		Metric Units	(model input)		
Source ID	Ht (ft)	Diam (in)	Temp (F) [2]	cfm	acfm	Ht (m)	Diam (m)	Temp (K)	Vel (m/s)
AN	68	16.0	575	1000	1961	20.7	0.4	575	7.13
BCD	68	24.0	575	2500	4902	20.7	0.6	575	7.93
E	68	34.0	575	8000	15687	20.7	0.9	575	12.64
Q	68	32.0	575	1000	1961	20.7	0.8	575	1.78
RS	82	72.0	80	100000	102335	25.0	1.8	300	18.39
L	68	22.0	575	3000	5883	20.7	0.6	575	11.32
К	68	16.0	575	1000	1961	20.7	0.4	575	7.13
G	93	34.0	80	28000	28654	28.3	0.9	300	23.09
Р	93	34.0	80	28000	28654	28.3	0.9	300	23.09
J	68	16.0	575	1000	1961	20.7	0.4	575	7.13
HF	83	32.0	575	1000	1961	25.3	0.8	575	1.78
M	68	22.0	575	3000	5883	20.7	0.6	575	11.32

[1] Stack parameter data from 1998 / 2000 facility data.

[2] 575 F assumed for abators, 300 F assumed for dilution stacks.





WRPLOT View - Lakes Environmental Software

# **BENNINGTON VT**





Attachment A-1 to Appendix A (Air Deposition Modeling)

PFOA Emissions from the Glass Fabric Coating Process

### Attachment A-1, PFOA Emissions from the Glass Fabric Coating Process

#### The Glass Fiber Fabric Coating Process:

In order to understand how PFOA is emitted, background information on the production process and the coating materials is helpful. Coating towers are used to apply and cure PTFE in the coated glass fiber fabric manufacturing process. The components of the coating towers include coating, drying, baking and fusing (sintering) sections. An annealing section sometimes follow the sintering section; it is used to control the coated fabric cooling rate to prevent it from cooling too quickly. A coating system also has mechanical systems for feeding the glass fiber fabric from the payout roll into the coating system, a take up roll to collect the coated fabric exiting the coating tower and devices for controlling the coating thickness, calendaring and trimming the coated fabric.

In the coating step, a dip tank coating system is used to apply a coating mixture to the glass fiber fabric. The glass fiber fabric comes off the payout roll and then through a series of rollers which direct the fabric down into the coating tank, up and out of the tank and then through adjustable rollers to control the coating thickness. A header tank feeds fresh coating into the dip tank to maintain the proper volume of coating in the dip tank.

The coating mixture is called a dispersion. Dispersions are mixtures of PTFE particles, surfactants and water. The PTFE particles are approximately 0.25 microns in diameter. APFO and other materials are utilized as surfactants in the dispersions. Surfactants are added to the dispersions to suspend the small PTFE particles in water so that the particles are evenly dispersed throughout the coating mixture. When APFO is added to a dispersion solution, it dissociates into PFOA and ammonium ions. The PFOA ions attach to the PTFE particles to hold them in "suspension". This allows the PTFE particles to be applied to the glass fiber fabric in a uniform coating.

After the coating is applied to the glass fiber fabric, the fabric passes through a coating tower which includes drying, baking and fusing (sintering) sections. The drying section is used to remove (evaporate) the water from the suspension coating on the fiber glass. The baking section is used to remove the surfactants from PTFE film. Fusing zone temperatures are sufficient to melt the PTFE particles so that a continuous PTFE film is formed. This is done to cure the PTFE film and ensure that it is firmly bonded to the fiber glass fabric. The coating tower designs have exhaust vents on each zone. The individual exhaust streams are then routed to a common duct. At the Merrimack, NH plant the combined exhaust is discharged to atmosphere through a stack. When they were in operation (1969 through 1978), the coating towers at the former plant on Northside Drive in Bennington, VT plant were also vented directly to atmosphere. At the former North Bennington, VT plant on Water Street (1978 through 2002), the exhaust was routed to an abator system for emission control before being vented to atmosphere. The abators were designed with thermal and catalytic oxidation sections. Typical temperatures in the coating tower are as follows:

- Drying zone: 95° to 150° C (200° to 300°F)
- Baking zone: 290° to 350° C (550° to 650°F)

- Sintering zone: 315° to 370° C (600° to 700°F)
- Abator Inlet: 135° to 150° C (275° to 300°F)
- Abator Operating Temperature: 304° to 316° C (580° to 600°F)

Natural gas burners are used to supply the heat necessary to achieve the temperatures needed in each zone of the tower.

#### APFO/PFOA Chemistry in the Coating Tower Drying and Bake Zones

As noted above, when the APFO is added to the suspension solution, it dissociates into PFOA and ammonium ions. As the glass fiber fabric passes through the drying zone, the water in the dispersion evaporates, and as the suspension solution concentrates, PFOA cations recombine with anions in the solution to form PFOA salts. Most of the PFOA ions will recombine with the ammonium ions to reform APFO, but some PFOA molecules may combine with other available anions resulting in the formation of other salts or the acid (PFOA).

When the glass fiber fabric passes into the bake zone, the temperature is raised to sufficient levels to drive the surfactant residuals out of the PTFE film. The types of compounds emitted from surfactant degradation in the bake zone depends upon whether PFOA ions in the suspension have re-combined into the ammonium salt (APFO) or have reacted with a hydrogen ion to form an acid (PFOA). APFO degrades readily at bake zone temperatures, and no PFOA is emitted from this reaction. PFOA degrades at a slower rate than APFO at baking zone temperatures, and the PFOA boiling point is below bake zone operation temperatures. So, PFOA which has not degraded can be vaporized and released from the film as a gas and exit the bake zone in the exhaust gas. These reactions are described in the section "Thermal Degradation of APFO and PFOA" later in this attachment.

#### Merrimack, NH Coating Tower Tests to Determine APFO Partition Factors

An industry trade group sponsored testing of PFOA emissions from glass fiber fabric coating operations (Society of Plastics Industry, 2005, 2005 Dispersion Processors Mass Balance (DPMB, 2005) Evaluation). DPMB evaluations were conducted on two types of coating tower designs at the Merrimack facility. All glass fiber fabric cloth coating towers at Bennington had the same design as glass fiber fabric coating towers tested at Merrimack.

In order to understand the Merrimack test results better, it is helpful to understand the two glass fiber fabric PTFE coating tower designs that were tested:

- An "up and out" tower is designed with the drying, baking and sintering zones stacked in a vertical arrangement. The glass fiber fabric enters at the bottom of the tower and exits at the top of the tower.
- An "up and down" tower is has a side by side arrangement. One side of the tower has the drying and baking zones and the other has the sintering zone and in some cases a tempering (annealing) zone. The glass fiber fabric enters at the bottom of the drying zone, passes up through the baking

zone, and then it is guided over rollers to the other tower section where it passes down through the sintering and tempering zones.

In either design, the drying, baking and sintering zones have the same operating temperatures and each zone has a separate exhaust.

- Drying zone: 95° to 150° C (200° to 300°F)
- Baking zone: 290° to 350° C (550° to 650°F)
- Sintering zone: 315° to 370° C (600° to 700°F)

For diagrams of both coating tower designs, see Attachment A-1-B - Figures 9.9 and 9.10 (Ebnesajjad, 2000), and chapter six in Drobny, 2001.

The tests conducted on PTFE coating towers as reported in DPMB, 2005 include "Process 1" which has an "up and out" design and "Process 2" which has an "up and down" design. A mass balance was conducted to track the fate of APFO through the production process on Process 1 and Process 2. The testing program included sampling and use rate measurements of APFO in the PFTE dispersions used to coat the glass fiber fabric, waste water and solid wastes generated in the production process, trimming waste from the coated fabric and stack testing on the combined coating tower exhaust gas streams. Air emissions testing was also performed on the drying section exhaust stream in Process 1; negligible amounts of PFOA were detected in the drying zone exhaust. The Merrimack coating tower exhaust gas streams did not have any add-on emissions controls like the abators used at the North Bennington plant. So the measured emission rates at Merrimack are representative of uncontrolled emissions at North Bennington. In Section 7 of DPMB, 2005, APFO air partition factors of 8.8% were reported for Process 1 (up and out) and 11% for Process 2 (up and down). The air partition factors represent the percentage of total APFO input used during the test which were measured in the air emissions. Tests on the coating tower exhaust stream measured PFOA emission rates, and emission rates were converted to the APFO equivalent for the mass balance calculations. The report noted that 87% of the APFO input to the production process was not detected in the environmental media tested. The report assumed that this material was either destroyed or remained in the final product. Attachment A-1-B, Figures 1 and 2 (DPMB, 2005) are process flow charts of Process 1 and Process 2 respectively. These figures document the production rates, process operating temperatures and residence times in Process 1 and Process 2 during the tests.

Testing in June 2005 on the MP Tower at the Merrimack facility (Barr, 2005) also showed an 11% partition factor when utilizing APFO dispersions similar to those used at North Bennington. The MP Tower is an "up and down" design like "Process 2" in DPMB, 2005.

The coating towers at the North Bennington plant which produced PTFE coated glass fiber fabric 1 meter in width were "up and out" designs, so the 8.8% partition factor was used to estimate uncontrolled PFOA emissions from these sources.

The coating towers at the North Bennington plant which produced PTFE coated glass fiber fabric 2 to 4 meters in width were "up and down" designs, so the 11% partition factor was used to estimate uncontrolled PFOA emissions from these sources.

#### **Thermal Degradation of APFO and PFOA**

The partition factors for APFO/PFOA emissions in the Merrimack tests can be explained by the decomposition rates and phase changes of APFO and PFOA which occur in the coating tower bake zones as summarized below. Attachment A-1-C, Figure 1 "Fate of APFO/PFOA in N Bennington, VT Coating Towers" is a coating tower flow diagram which shows the fate of APFO and PFOA as they pass through the coating tower sections and the abator.

The following literature sources describe the thermal decomposition of APFO and PFOA to characterize chemical reactions that occur in the coating tower bake zones:

- Gas-Phase NMR Technique for Studying the Thermolysis of Materials: Thermal Decomposition of Ammonium Perfluorooctanoate (Krusic. et al., 2004)
- Gas-phase NMR studies of the Thermolysis of Perfluorooctanoic Acid (Krusic. et al., 2005)

The authors of these papers conducted experiments to quantify the thermal degradation rates of APFO and PFOA. The experiments were done by placing APFO and PFOA samples in glass vials and heating up the samples to temperatures at which APFO and PFOA would thermally degrade. In some cases, degradation temperatures exceeded the limits of the measurement device. The contents of the vials were tested before and after exposure to the temperatures necessary to achieve thermal decomposition. A Nuclear Magnetic Resonance (NMR) analyzer was used to measure the concentrations of the APFO, PFOA and fluorocarbon degradation products. APFO readily decomposed under the experimental conditions. Pure samples of PFOA decomposed slowly with half-lives of several minutes. Decomposition rates for PFOA were shown to decrease significantly by increasing the amount of PFOA in the sample vial, adding water to the sample vial or by increasing the available surface area for degradation reactions to occur on by adding crushed glass to the sample vials.

APFO thermal decomposition occurs as a first order reaction with a half-life of a few seconds at bake zone operating temperatures. The authors of the first reference developed a correction for predicting the PFOA first order decomposition reaction rates at various temperatures. This correlation was used to project APFO degradation rates at bake zone operating temperatures. APFO is an unstable liquid and decomposes before it can be vaporized. Table 1-1 below lists the physical properties of both chemicals. APFO degrades into 1H-perfluoroheptane, carbon dioxide and ammonia as follows:

$$CF_3(CF_2)_5CF_2CO_2 \cdot NH_4 \rightarrow CF_3(CF_2)_5CF_2H + CO_2 + NH_3$$

PFOA decomposes into two types of fluorocarbon molecules releasing carbon dioxide and hydrogen fluoride as byproducts. The decomposition rates are slower, and can be approximated by first order reaction rates, but do not correlate as well to first order kinetics as APFO does. The reactions are as follows:

a)  $CF_3(CF_2)_5CF_2COOH \rightarrow CF_3(CF_2)_5CF_2H + CO_2$  or

#### b) $CF_3(CF_2)_5CF_2COOH \rightarrow CF_3(CF_2)_4CF_2=CF_2 + CO_2 + HF$

PFOA is a stable liquid and its boiling point is at a lower temperature than bake zone operating temperatures; so, it can be released from the PTFE film in gaseous form in the bake zone.

	Meltir	ng Point	Boiling	Point	Vapor Press	H₂O Sol @25°C
	°C	°F	°C	°F	Pa@ 25° C	g/L
PFOA	54.3	129.7	188.0	370.4	4.2	9.5
APFO	161.0	321.8	NA Decon	nposition	0.0081	>500
APFO Decomposition starts at:			130.0	266.0		

Table 1-1 Com	oarison of Ph	vsical Proper	ties PFOA an	d APFO
		,		

1. UNEP/POPS/POPRC.1/5, 2015

2. US EPA, 2016

Table 1-2 and Table 1-3 on the following page present a comparison the PFOA boiling point to the coating tower zone temperature and the APFO thermal degradation half-life and percent removal rate. The APFO half-life (Krusic. et al. 2004) and percent removal rates calculated for the conditions for Process 1 and Process 2 testing at Merrimack from the DPMB reports as listed in Attachment A-1-B Figures 1 and 2. Both Tables show that zone temperatures do not exceed PFOA boiling point in the drying zones. This is consistent with the drying zone test results for Process 1 that showed negligible amounts of PFOA in the drying zone exhaust.

The bake and fuse (sinter) zones of both processes were above the PFOA boiling point. So it possible that PFOA in the PTFE film is vaporized in the bake and fuse zones and exhausted from the coating tower in the tower exhaust gas.

Both tables show that nearly 100% of APFO degrades in the bake zones of each tower. APFO degradation rates are so fast under bake zone conditions that even at the shorter 0.3 minute residence time in Process 1 nearly all APFO is destroyed. These tables show that:

- The low APFO partition rates in both processes demonstrate that the majority of PFOA ions in the dispersion solution are associated with ammonium ions in the drying zones and the APFO salts are destroyed in the bake zone.
- Process conditions are such that nearly complete destruction of APFO occurs in the bake zone regardless of whether the tower is an "up and out" or "up and down" design.
- Process conditions in the bake zone are hot enough to vaporize PFOA reporting to the bake zone from the drying zone, thus providing a pathway for PFOA to leave the coating tower in the bake zone (or fuse zone) exhaust streams.

	> PFOA BP?	T° F	T° C	Т° К	k min <sup>-1</sup>	t <sub>1/2</sub> min	t <sub>1/2</sub> Sec	λ	Residence time, sec	% APFO Removed
Durving	No	250	121.1	394.3	9.48E-06	7.31E+04	4.39E+06	1.58E-07	18	2.84E-04
Zone	No	275	135.0	408.2	4.66E-05	1.49E+04	8.93E+05	7.76E-07	18	1.40E-03
20112	No	300	148.9	422.0	2.06E-04	3.36E+03	2.02E+05	3.44E-06	18	6.19E-03
	Yes	550	287.8	560.9	10.8	0.06	3.8406	0.18	18	96.12
Bake Zone	Yes	575	301.7	574.8	24.1	0.03	1.7230	0.40	18	99.93
	Yes	600	315.6	588.7	51.8	0.01	0.8023	0.86	18	100.00
<b>-</b>	Yes	600	315.6	588.7	51.8	0.01	0.8023	0.86	18	100.00
Fusing	Yes	625	329.4	602.6	107.5	0.01	0.3868	1.79	18	100.00
20110	Yes	650	343.3	616.5	215.9	0.003	0.1926	3.60	18	100.00

Table 1-2 APFO Half Life vs Temperature Process 1 - Partition Factor = 8.8%

#### Table 1-3 APFO Half Life vs Temperature Process 2 - Partition Factor = 11%

	> PFOA BP?	T° F	T° C	T° K	k min <sup>-1</sup>	t <sub>1/2</sub> min	t 1/2 Sec	λ	Residence time, sec	% APFO Removed
Durving	No	200	93.3	366.5	2.75E-07	2.52E+06	1.51E+08	4.58E-09	90	4.12E-05
Zone	No	225	107.2	380.4	1.72E-06	4.03E+05	2.42E+07	2.87E-08	90	2.58E-04
20112	No	250	121.1	394.3	9.48E-06	7.31E+04	4.39E+06	1.58E-07	90	1.42E-03
	Yes	550	287.8	560.9	10.8	0.06	3.8406	0.18	90	100.00
Bake Zone	Yes	600	315.6	588.7	51.8	0.01	0.8023	0.86	90	100.00
	Yes	650	343.3	616.5	215.9	0.00	0.1926	3.60	90	100.00
<b>F</b>	Yes	650	343.3	616.5	215.9	0.00	0.1926	3.60	90	100.00
Fusing	Yes	675	357.2	630.4	420.8	0.00	0.0988	7.01	90	100.00
20116	Yes	700	371.1	644.3	797.0	0.001	0.0522	13.28	90	100.00

PFOA has a slower thermal degradation rate than PFOA, but insufficient information exists to project degradation rates in the coating towers. Half-lives for PFOA (Krusic. et al., 2005) are listed in Table 1-4 below. However, these results are for PFOA in an inert environment.

T° F	T° C	k min⁻¹	t <sub>1/2</sub> min	
671.0	355.0	5.00E-03	138.6	
698.0	370.0	9.30E-03	74.5	Fuse Zone Operating Temperature
725.0	385.0	2.64E-02	26.3	

Table 1-4 PFOA Pseudo Half Life vs Tempe	erature in an Inert Environment
--	---------------------------------

Additional experiments by the authors showed that PFOA degradation rates could be improved by increasing the amount of sample in the test vial, adding water to the test vial and adding crushed glass to the test vial. The shortest reported half-life for these experiments was 1.3 hours at 307° C in the presence of crushed sodium borosilicate glass. While these experiments show that there is potential for improving PFOA degradation rates by altering the experimental conditions, test conditions did not mimic actual conditions in a coating tower and no correlation was developed which could be utilized for this project.

#### Abator PFOA Control:

The North Bennington, VT coating towers were equipped with "abators" for controlling VOC emissions from the towers. The abators were thermo-catalytic control devices which provided 46% control efficiency for PFOA. The mechanism for PFOA destruction is:

- 1. PFOA adsorbs onto particles in the coating tower exhaust duct.
- 2. As particulates are destroyed in the abators, PFOA absorbed on the particulates is also destroyed.

This control efficiency is supported by the following:

- a) PFOA readily sorbs onto particulate matter, and PFOA in ambient air is largely bound to particulate matter (US EPA, 2016).
- b) The coating tower exhaust contains particulate matter which adsorbs PFOA. Particulate matter is generated in the bake and fuse zones of a coating tower
- c) PFOA emissions can be captured in particulate control devices. June 2005 testing of the MA tower in Merrimack, NH (Barr, 2005) had a 98% PFOA removal rate from a PFOA control device which was designed on particulate control methodologies (condensation by water quenching and filtration)
- d) The PFOA degradation experiments (Krusic, et al., 2005) showed an increase in PFOA degradation rates with:

- i. Addition of water
- ii. Increasing surface area for reactions by addition of crushed glass

Decomposition of carboxylic acid vapors is prone to heterogeneous effects and surface assistance. The addition of quartz crushed glass was tested to verify that these effects would accelerate the decomposition of PFOA. Therefore, a similar increase decomposition rates of PFOA is likely to occur when PFOA is sorbs onto particulate matter prior to entering the abators. The addition of water vapor from the drying zone to the abator may also improve PFOA decomposition rates.

e) Testing of the R abator at North Bennington, VT showed a 46% reduction in particulate emissions with the abator and catalyst in service versus operation with the abator turned off (TRC Environmental Corporation, 1999).

Therefore, it is reasonable to conclude that the R Abator test results are indicative of the abators' PFOA control efficiency.

### References

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- UNEP (United Nations Environment Programme)/POPS/POPRC.1/5, 9 June 2015. "Proposal to list pentadecafluorooctanoic acid (CAS No: 335-67-1, PFOA, perfluorooctanoic acid), its salts and PFOArelated compounds in Annexes A, B and/or C to the Stockholm Convention on Persistent Organic Pollutants
- United States Environmental Protection Agency (US EPA), 2016, "Drinking Water Health Advisory for Perfluorooctanoic Acid (PFOA)" EPA 822-R-16-005, May 3, 2016.

# Attachment A-1-A to Appendix A (Air Deposition Modeling)

Figures 9.9 and 9.10 from Chapter 9, Fabrication and Processing of PTFE Dispersions



Figure 9.8 Simple counter-current tower.<sup>[9]</sup>

Countercurrent Flow Each Zone or Direct Impingement Slot Dryers or Radiant Heat:—Separate Exhausts Each Zone

Countercurrent Flow Each Zone or Direct Impingement Slot Dryers or Radiant Heat:—Separate Exhausts Each Zone



Figure 9.9 Modern coating towers.<sup>[9]</sup>

Figure 9.10 Modern coating towers.<sup>[9]</sup>

# Attachment A-1-B to Appendix A (Air Deposition Modeling)

Figures 1 and 2, DPRM, 2005 Attachment 2



Process 1+2 Flow Charts/Process 1



Process 1+2 Flow Charts/Process 2

# Attachment A-1-C to Appendix A (Air Deposition Modeling)

Figure 1 Fate of APFO/PFOA in Saint Gobain Performance Plastics Coating Towers

#### Figure 1 Fate of APFO/PFOA in N Bennington, VT Coating Towers





# Attachment A2

Annual Simulated Air Deposition: 1969-2001











YEAR: 1969 AIR DEPOSITION OF PFOA



# 0 0.25 0.5 Miles

BARR

# PFOA Deposition g/m²/yr



1.1e-05 - 1.8e-05
1.9e-05 - 3.2e-05
3.3e-05 - 5.6e-05
5.7e-05 - 1.0e-04
1.1e-04 - 1.8e-04
1.9e-04 - 3.2e-04
> 3.2e-04

YEAR: 1970 AIR DEPOSITION OF PFOA





2001

016-

BARR

# PFOA Deposition





YEAR: 1971 AIR DEPOSITION OF PFOA











YEAR: 1972 AIR DEPOSITION OF PFOA





PFOA	Deposition





YEAR: 1973 AIR DEPOSITION OF PFOA





2001

016-

BARR

# PFOA Deposition





YEAR: 1974 AIR DEPOSITION OF PFOA











YEAR: 1975 AIR DEPOSITION OF PFOA





# PFOA Deposition





YEAR: 1976 AIR DEPOSITION OF PFOA









YEAR: 1977 AIR DEPOSITION OF PFOA





PFOA [	Deposition
--------	------------





YEAR: 1978 AIR DEPOSITION OF PFOA











YEAR: 1979 AIR DEPOSITION OF PFOA



BARR

# g/m²/yr





YEAR: 1980 AIR DEPOSITION OF PFOA









YEAR: 1981 AIR DEPOSITION OF PFOA









YEAR: 1982 AIR DEPOSITION OF PFOA



# 0 0.25 0.5 Miles

BARR







YEAR: 1983 AIR DEPOSITION OF PFOA











YEAR: 1984 AIR DEPOSITION OF PFOA









YEAR: 1985 AIR DEPOSITION OF PFOA







1.1e-05 - 1.8e-05 1.9e-05 - 3.2e-05 3.3e-05 - 5.6e-05 5.7e-05 - 1.0e-04 1.1e-04 - 1.8e-04 1.9e-04 - 3.2e-04 > 3.2e-04

YEAR: 1986 AIR DEPOSITION OF PFOA









YEAR: 1987 AIR DEPOSITION OF PFOA









AIR DEPOSITION OF PFOA



0 0.25 0.5 Miles

BARR



1.1e-05 - 1.8e-05 1.9e-05 - 3.2e-05 3.3e-05 - 5.6e-05 5.7e-05 - 1.0e-04 1.1e-04 - 1.8e-04 1.9e-04 - 3.2e-04 > 3.2e-04

YEAR: 1989 AIR DEPOSITION OF PFOA











AIR DEPOSITION OF PFOA



BARR





3.3e-05 - 5.6e-05 5.7e-05 - 1.0e-04 1.1e-04 - 1.8e-04 1.9e-04 - 3.2e-04 > 3.2e-04

AIR DEPOSITION OF PFOA









AIR DEPOSITION OF PFOA











YEAR: 1993 AIR DEPOSITION OF PFOA



5.7e-05 - 1.0e-04

1.1e-04 - 1.8e-04

1.9e-04 - 3.2e-04

> 3.2e-04

North Bennington, VT Saint-Gobain



0.5

0.25

Miles

3.3e-06 - 5.6e-06 5.7e-06 - 1.0e-05









YEAR: 1995 AIR DEPOSITION OF PFOA



BARR



3.3e-05 - 5.6e-05 5.7e-05 - 1.0e-04 1.1e-04 - 1.8e-04 1.9e-04 - 3.2e-04 > 3.2e-04

AIR DEPOSITION OF PFOA







YEAR: 1997 AIR DEPOSITION OF PFOA









AIR DEPOSITION OF PFOA



BARR



3.3e-05 - 5.6e-05 5.7e-05 - 1.0e-04 1.1e-04 - 1.8e-04 1.9e-04 - 3.2e-04 > 3.2e-04

AIR DEPOSITION OF PFOA



ir M

2016-

BARR





AIR DEPOSITION OF PFOA



ir M

2016-

BARR



3.3e-06 - 5.6e-06 5.7e-06 - 1.0e-05

3.3e-05 - 5.6e-05 5.7e-05 - 1.0e-04 1.1e-04 - 1.8e-04 1.9e-04 - 3.2e-04 > 3.2e-04

AIR DEPOSITION OF PFOA