

An Investigation of Alternative Tracer Gases and Computational Methods for Measuring Leakage in High Moisture Content Gaseous Filtration Systems

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ABSTRACT

This paper examines the suitability of several tracer gases for use in performing in-place leak tests of gaseous filtration systems per ANSI/ASME N510, N511, and AG-1. The tracer gases investigated were trichlorofluoromethane (R-11), 1,1,1,2,2,3,4,5,5,5-decafluoro-pentane (Vertrel-XF), perfluoromethylcyclopentane (PMCP), perfluoromethylcyclohexane (PMCH), and perfluorodimethylcyclohexane (PDCH).

R-11 is currently the most common tracer used in measuring the mechanical leakage in a nuclear gaseous filtration system. However, it is an ozone depleting substance regulated by the Montreal Protocol. Vertrel-XF, commonly referred to as "Vertrel", was chosen by the US military to test their gaseous filtration systems because it is not an ozone depleting substance. The perfluorocarbon tracers are also non-ozone depleting substances, are easily detected by traditional electron capture gas chromatography, and are suitably retained on carbon filters for performance of the in-place leakage test.

The effect of each tracer on radioiodine efficiency of nuclear grade carbon was measured. Consecutive measurements of mechanical leakage of a small gaseous filtration system were made under moisture loadings from less than 5% to 27% by weight. All of the tracers showed good agreement in the calculation of mechanical leakage at less than 17% moisture loadings. Above this level, R-11, PMCP and PMCH all exhibited various degrees of desorption from the carbon that made the calculation of mechanical leakage difficult or impossible. At high moisture loadings of approximately 25%, Vertrel exhibited moderate desorption but calculation of mechanical leakage was still possible using the technique of estimating leakage by extrapolating the desorption curve back to the time at which the upstream concentration stabilized. This technique is not adequately described in more recent versions of ASME N510, N511, and AG-1 but is relied on by test personnel, and included in many nuclear plant procedures based on descriptions of the technique appearing in earlier versions of ANSI/ASME N510. PDCH performed the best at these high moisture loading levels and exhibited minimal to no desorption from the carbon.

INTRODUCTION

In-place filter testing is an integral part of maintaining safety related ventilation systems. There are two tests for mechanical leakage that may be required by an in-place filter test program depending on the system design. Leakage tests of the HEPA filter bank are performed using submicron particles, while leakage tests of carbon adsorber banks are performed using a tracer gas. Both tests measure bypass of the filter or adsorber banks. Results are reported in percent mechanical leakage, expressed as a percentage of total system flow rate. Bypass leakage may be caused by a variety of conditions including; manufacturing defects of the filter housing, mounting frame seating issues, media damage in the case of HEPA filters, and packing density issues in the case of carbon adsorbers. This paper focuses on the challenge gases or vapors (tracers) that may be used to test bypass leakage around the carbon filters. The three main points of discussion are:

1. Comparison of traditional and alternative tracers
2. Effects of carbon bed moisture loading on test results
3. Alternative calculation methods to account for breakthrough

First, we will discuss the suitability of Vertrel and Perfluorocarbon Tracers (PFTs) as surrogates for R-11. New carbon beds at Savannah River Laboratory (SRL) were originally tested for leaks using dichlorodifluoromethane (R-12) as the tracer in the early 1960s while installed carbon beds subsequently used 1,1,2,2-tetrachloro-1,2-difluoroethane (R-112) as the tracer [9,10]. The technical basis for the carbon adsorber bypass test is described in a number of progress reports issued by Savannah River Laboratory (SRL) in the 1960's titled "Nondestructive Test of Carbon Beds for Reactor Confinement Applications" [9,10,11]. In 1971, the ANSI N45.8.3 Task Group recommended that R-11, rather than R-112 be used for non-destructive testing of carbon beds due to its lower detection limit, lower boiling point (easier handling, generation, and purging of the carbon bed) and the discontinued manufacture of R-112 by Dupont. Currently, US nuclear power plant Air Filtration Unit (AFU) testing programs use R-11 exclusively. Due to ozone depletion concerns most ChemBio AFU testing programs are not allowed to use R-11 and have adopted Vertrel as their tracer gas of choice. Other testers, including the authors, have utilized perfluorocarbon tracers (PFTs) to test ChemBio AFUs.

Secondly, we will compare the desorption (or breakthrough) curves for the various tracers at different levels of moisture loading. The SRL papers focused on using the less volatile R-112 as a surrogate for R-12 because the R-12 technique was limited to use in "carbon containing no more than 5% sorbed H₂O" and carbon in use had "adsorbed impurities that cause the R-12 to desorb too quickly" [8, p. 5]. Depending upon the physical characteristics of the tracer used (typically molecular weight and boiling point) the negative effects of moisture on the desorption of tracer during a bypass leakage test can be limited. Due to the unavailability of R-112, the lower detectability limit, ease of handling, and better inlet generation precision of R-11, the industry moved to R-11 as a substitute for R-112. The move to R-11 was not without its disadvantages, principally sacrificing the excellent retention of R-112 on carbon for ease of handling, generation and detection. The industry is now interested in moving toward alternative tracer gases that incorporate the best characteristics of R-112 and R-11 for the in-place leak test while at the same time assuring candidate tracers do not adversely affect the radioiodine removal characteristics of the carbon.

Finally, alternative calculational methods for bypass leakage determination using exponential, square, and quadratic curves will be presented and compared to a strict averaging method. The original work outlined in the SRL DP-1082 mentions that the adsorption efficiency “curve can be extrapolated back to time zero with confidence to determine leakage flow...” [10, p. 21]. To determine the leakage flow around carbon beds ASME N510-2007 states that the “adsorber bank leak rate is determined from the ratio of downstream to upstream concentration” [5, section 10.2]. The vague instructions contained in ASME N510 could result in test personnel averaging downstream concentrations which will *overestimate* the leakage flow when breakthrough occurs.

ALTERNATIVE TRACERS

Physical properties of a tracer, such as molecular weight (MW) and boiling point (BP), determine its effectiveness for use in testing bypass leakage on installed carbon AFUs. Environmental criteria may also be important such as the ozone depletion capabilities and global warming potential (GWP). Additionally, cost is a major factor especially if a large number of AFUs must be tested and retested on a periodic basis. One often overlooked property of a tracer gas are the problems associated with obtaining quality calibration standards at the downstream measurement Limit of Detection (LOD). All of the tracer gases used during this testing were referenced to calibration standards prepared and analyzed by the same independent laboratory. While there is no “perfect” tracer, the physical properties, filter test requirements, environmental concerns, and cost should all be considered for each individual testing program. A summary of the properties of various tracer gases is presented Table 1 below.

Table 1

Properties of historical and currently used tracers for gaseous filter leak testing

Tracer	MW	Boiling Point (C)	Vapor Pressure @ 25C (mm Hg)	ECD-GC LOD (ppb)	Ozone Depl.?	Global Warming Potential (20 yr)	Cost (\$/kg)	Cal. Gas Uncertainty (0.1ppb / 1.0ppb)
R-12	121	-29.8	4390.6	??	Yes	8100	\$130	N/A
R-112	204	92.5		??	Yes	~5000	N/A	N/A
R-11	137	23.8	801.3	<0.1	Yes	3800	\$35	N/A / 5%
Vertrel	252	55	234.7	~0.4*	No	4140	\$55	N/A / 20%
PMCP	300	48.0	337.5	0.05	No	~5000-8000	\$450	20% / 5%
PMCH	350	76.0	105.8	<0.1	No	~5000-8000	\$450	20% / 5%
PDCH	400	102	36.0	0.05	No	~5000-8000	\$300	20% / 5%

* - Vertrel LOD estimated based on 1 ppb ECD-GC response and quality calibration standards cannot be made below 0.5 ppb

R-12 and R-112 have not been used for filter testing for many decades and their data were presented for comparison only. The longest used tracer, R-11, has many good qualities in that it is inexpensive, has excellent ECD-GC response, and quality low level calibration gases can be made. The negatives are that it is an ozone depletor subject to the Montreal Protocol and it

has poorer retention on carbon due to its lower molecular weight and boiling point. R-11's most utilized substitute, Vertrel is inexpensive, has the environmental benefits of not being an ozone depletor, and also exhibits a low GWP. It is also better retained on carbon due to its higher MW and boiling point. However, Vertrel has limitations in that the LOD is a factor of 5 to 10 higher than the other tracers. Additionally low level calibration gases are subject to much higher uncertainties due to Vertrel's response in the analysis technique and quantification methods of the gas manufacturer. To combat this, Vertrel needs to be used at higher concentrations which will offset some of its environmental benefits. As for the PFTs, the main drawbacks are their cost and higher GWPs. PFTs main benefits are great ECD-GC response and the ability to obtain quality low level calibration gases. The authors prefer PDCH because its retention time on carbon is the best of any of the tracers. In addition, it is also slightly less expensive than the other PFTs.

Per Non-Mandatory Appendix TA-C of AG-1, *Challenge Gas and Aerosol Substitute Selection Criteria*, when compared to R-11, R-12, R-112, or R-112a, alternative challenge gases (tracers) need to meet the following characteristics:

1. Give the same In-place Leak Test results
2. Have similar retention times on activated carbons, at the same concentration levels
3. Have similar lower detection limit sensitivity and precision in the concentration range of use
4. Exhibits chemical and radiological stability under the test conditions
5. Causes no degradation of the carbon and its impregnant(s) under the test conditions
6. Listed in the EPA's TSCA inventory for commercial use if it is toxic

One purpose of this study was to show that the alternative tracers satisfy criterion #1 plus, not only satisfy criterion #2, but actually improve upon the retention time for higher moisture loaded carbon. The comparison filter tests performed show that all of the alternative tracers satisfy criterion #1 by returning the same leak test results within the uncertainty level of the technique. Additionally, the tests performed at various moisture loadings show that alternative tracers not only satisfy criterion #2 - similar retention levels - but some can actually be an improvement from R-11 for high moisture loaded carbon.

All of the PFTs satisfy criterion #3 with one caveat. Vertrel does have lower levels of precision in the manufacture of calibration gases at the sub-ppb level. However, above 1 ppb the uncertainties are acceptable and therefore satisfy criterion #3 in its range of use. All of the tracers studied can be analyzed by an electron capture gas chromatograph (ECD-GC). An ECD-GC utilizes the high electron affinity of gases or vapors with halogen group elements to provide a measurable signal. Refrigerants such as R-11, Vertrel, and various perfluorocarbons are such gases.

All of the alternative tracers are non-toxic, stable, and have no known chemical or radiological incompatibilities that would be encountered under normal test conditions. As such, all of the alternative tracers satisfy criteria #4 and #6.

To satisfy criterion #5, Radioiodine Penetration / Efficiency tests were performed. New carbon impregnated with 5% triethylenediamine (TEDA) was tested both without tracer and with 1% of dry carbon weight loading for each tracer. The 1% loading was chosen as a conservative

screening tool. Mass loadings under actual field test conditions for single or multiple tests are well below the 1% tracer loading therefore we would then expect to see no degradation when used in the field. Within the uncertainty of the measurement, all of the alternative tracers studied exhibited the same % Penetration and % Efficiency for non-loaded and 1%-loaded new carbon. This shows that the alternative tracers will cause no degradation of the carbon or TEDA under test conditions. The test results are summarized in Table 2 below.

Table 2
Radioiodine Penetration / Efficiency test results

Sample	Date	% Penetration	% Efficiency	% Change
New Carbon with no PMCH	2/12/2016	0.35	99.65	
New Carbon with 1% PMCH	2/12/2016	0.31	99.69	+0.04%
New Carbon with no PDCH	2/16/2016	0.29	99.71	
New Carbon with 1% PDCH	2/16/2016	0.30	99.70	-0.01%
New Carbon with no Vertrel	3/10/2016	0.37	99.63	
New Carbon with 1% Vertrel	3/10/2016	0.35	99.65	+0.02%

A cautionary note for Vertrel is that per the MSDS, Vertrel “is incompatible with strong bases”. TEDA is a basic amine but apparently not a strong enough base, like KOH or NaOH, to exhibit degradation when exposed to 1% Vertrel. The test results provided above support this supposition.

TEST SET-UP

The breakthrough tests were performed on a filtration unit supplied by NCS Corporation. The AFU is a test bed utilized in their “In-Place Testing of Nuclear Air Cleaning Systems” training class. It is a small unit with a 12”x12” pre-filter, a 12”x12” HEPA filter, and a 12”x12”x2” carbon adsorber bank. For this testing, it was operated at approximately 60 m³/hr (35 cfm) which gave a residence time of 0.29 seconds. The carbon adsorber bank was loaded and reloaded from the same homogeneous batch of impregnated carbon for all of the tests.

After loading the carbon, the system fan was energized and moisture loading commenced. Moisture loading was varied by “tenting” the inlet of the AFU and setting a simple humidifier to a specified output. The humidifier was turned on the afternoon prior to the test allowing the carbon to equilibrate for approximately 16 hours prior to performance of a filter test. A picture of the test set-up is provided in Figure 1.

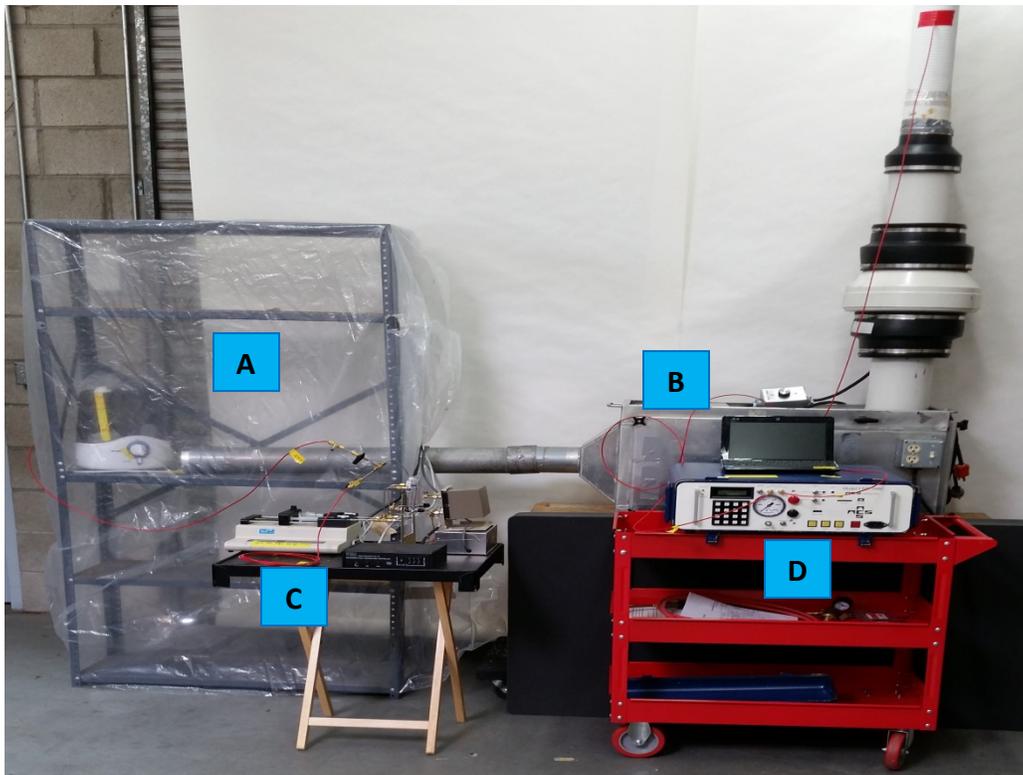


Figure 1 – Test set-up showing (A) humidifier in tent, (B) filter housing, (C) syringe pump injection system, and (D) LMP-3000 Halocarbon Monitor.

A syringe pump was utilized to accurately and consistently meter liquid tracer into a flowing, heated stream of nitrogen which was injected into a coiled section of 4" round ducting attached to the inlet of the AHU test bed. Test results confirm that each tracer was consistently challenged at the upstream sample location at a tracer concentration of approximately 10-15 ppm.

Filter test analyses were all performed on the same LMP-3000 Tracer Gas Monitor. The LMP-3000 as configured was capable of measuring tracer gas concentrations of approximately 1-30 ppm on the upstream sample port and sub-ppb concentrations on the downstream sample port for all tracers utilized in this study. Filter test performance for each tracer was performed sequentially. Note that concurrent testing cannot be performed without negatively impacting the sample throughput time of the LMP-3000 for earlier eluting tracers. Concurrent testing would also cause the breakthrough of early eluting tracers to negatively impact the analysis of later eluting tracers.

Two to four tracers were tested on the AFU each day at a particular moisture loading. Pre-test, challenge, and post-test sampling each lasted approximately 30 minutes. The separation column on the LMP-3000 distinctly separates each tracer gas though full separation of R-11 and PMCP is difficult to achieve. As such, the testing program sequenced the tracer challenges based upon the elution time of the tracer in the LMP-3000. Early eluting tracers like R-11 or PMCP were used to challenge the AFU first. After completion of the first test the LMP-3000 was optimized for the next challenge tracer. PMCH, being the middle eluting tracer, was challenged second while a late eluting tracer either Vertrel or PDCH was used for the last

challenge. Note that on two days Vertrel was used as a challenge after PDCH because PDCH was fully retained on the carbon.

Since carbon samples could not be obtained during a comparison test sequence, each day's testing must assume that the % moisture loading was at and maintained equilibrium throughout that test day. After the day's testing at a particular moisture loading, all of the "wet carbon" was removed and an approximate 250 ml sample was collected and weighed. This sample was then baked in an oven at 120°C overnight with a nitrogen purge and then weighed again the following morning. The % moisture loading was calculated differencing these two values and dividing by the initial "wet carbon" value.

TRACER PERFORMANCE AT VARIOUS MOISTURE LOADINGS

The adsorber in-place leak test takes advantage of the adsorptive characteristics of activated carbon and the temporary delay of tracer gas to measure mechanical leakage around or through an activated carbon bed. Leakage is defined as the percentage of system flow that is NOT treated by the system carbon bed. For low moisture conditions lower MW tracers like R-11 are retained on carbon for the duration of the test while very high MW tracers could, in theory, be permanently retained on the carbon.

Another objective of this study was to compare the performance of various tracers under different moisture loadings. This is a very important factor when performing in-place filter tests on carbon adsorber AFUs that have been in service for a period of time. Many locales are subject to high humidity conditions and seasoned test engineers know all too well the complications that arise when filter banks are subject to these high humidity conditions; the carbon is unable to retain the challenge tracer for the duration of the test. When the carbon adsorber is unable to retain the tracer this is termed desorption or "breakthrough". The degree of breakthrough is dependent upon carbon adsorber AFU conditions, namely air velocity, bed thickness, carbon type, relative humidity, and poisoning of carbon by adsorbed chemical compounds. Poisoning and moisture fill up a portion of the sites available on the carbon to retain chemicals or vapors, such as the tracers used to perform the in-place leakage test. Figure 2 below graphically represents an in-place leak test with a measureable mechanical leak coupled with moderate breakthrough, or desorption. Figure 3 illustrates the problem when there is significant breakthrough, limiting the ability to obtain sufficient data points to characterize the desorption curve.

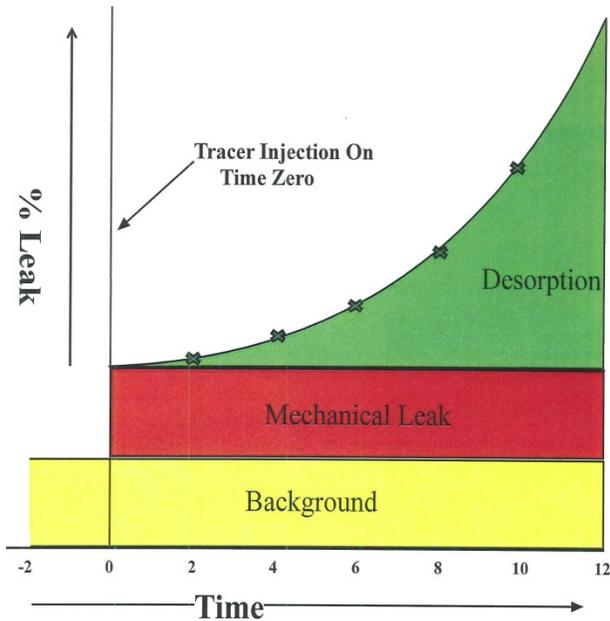


Figure 2 – Mechanical leak with moderate desorption

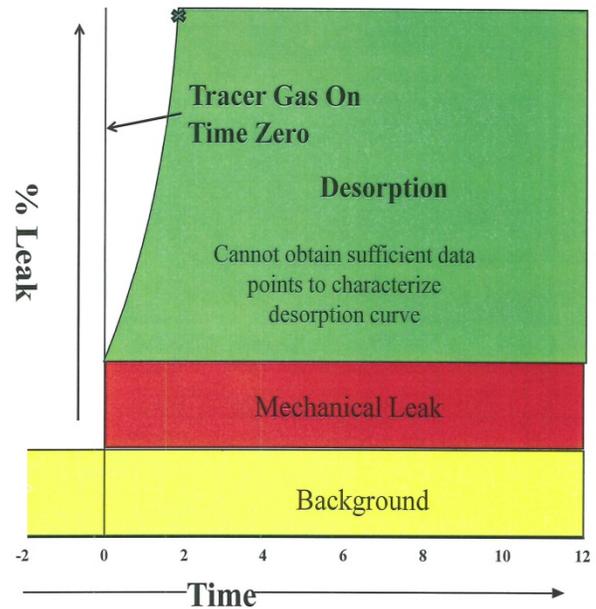


Figure 3 – Mechanical leak with rapid desorption

Selection of the proper tracer can mitigate some of the problems associated with in-place testing on AFUs with poisoned or moisture loaded carbon. Higher MW and higher BP tracers will be retained on the carbon for a longer period of time. As previously mentioned, R-12 could only be used to test new carbon with moisture loading <5% so it was not utilized for any in-place filter testing. R-11 appears to have limited breakthrough in our tests with moisture loadings up to ~15%. To perform an in-place filter test in the field above this moisture level, preconditioning (i.e. drying) was necessary prior to performing the test. Typically, drying would delay the performance of the in-place test for at least a day or two. Hence, alternative tracers were sought to limit the effect of moisture loaded carbon on the in-place filter testing program.

Based on the results of this investigation the use of R-11 and PMCP is significantly diminished at moisture loadings above approximately 20%. At moisture loadings of approximately 25%, PMCH also showed significant desorption but it was not as drastic as R-11 or PMCP which would be expected based off its higher molecular weight and boiling point. Additionally, against convention, Vertrel appears to perform better at higher moisture loadings than PMCH even though it has a lower molecular weight and boiling point. It should also be noted that while Vertrel exhibited moderate breakthrough at high moisture loadings (approximately 25%) the calculation of mechanical leakage based on a curve extrapolated back to time = 0 appears to be accurate. Because PDCH exhibited minimal to no breakthrough at moisture loadings up to at least 25% water content by weight, it appears to be ideal to use in high moisture content systems. A summary of the comparison tests performed at various moisture loadings is found in Table 3 and a plot of a comparison test performed on 1/27/2015 is presented in Figure 4. Note that even at approximately 15 minutes of challenge there was no breakthrough of PDCH.

Table 3

Summary of filter comparison test results at various moisture loadings

Calculated Leakage – Exponential								
Date	12/17/14	12/18/14	12/19/14	12/22/14	12/23/14	01/23/15	01/27/15	02/05/15
% Water	13.2%	16.4%	27.5%	24.5%	12.9%	16.2%	24.5%	24.6%
R-11 Leak %	0.066%		0.037%		0.049%		0.166%	
Breakthrough?	No		Yes		No		Yes	
PMCP Leak %		0.065%		11.02%*		0.095%		Overrange
Breakthrough?		No		Yes		No		Yes
PMCH Leak %	0.074%	0.053%	0.035%	0.51%*	0.038%	0.086%		1.22%*
Breakthrough?	No	No	No	Yes	No	No		Yes
PDCH Leak %	0.058%		0.037%		0.042%	0.084%	0.083%	
Breakthrough?	No		No		No	No	No	
Vertrel Leak %		0.051%		0.020%	0.040%	0.086%		0.063%
Breakthrough?		No		Yes	No	No		Yes

* – Leakage plus desorption, breakthrough too massive to accurately calculate the leak %

Note that two days of testing were omitted because the breakthrough for all tracers was too large (1/11/15) or the bypass leakage was too large (1/14/15) for effective calculational purposes.

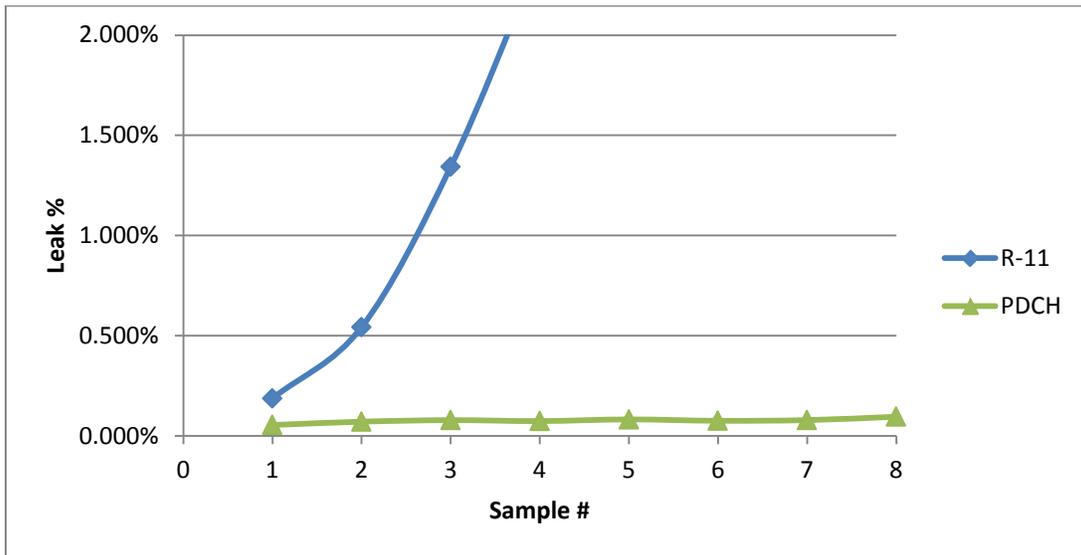


Figure 4 – Filter comparison test performed on 1/27/15, moisture loading = 24.5% by weight. R-11 calculated leak = 0.16% (overestimation), PDCH calculated leak = 0.08%.

ALTERNATIVE CALCULATIONAL METHODS TO ACCOUNT FOR LEAKAGE

The simple calculation of bypass leakage in carbon adsorber filtration units is outlined in AG-1 and Mandatory Appendix IV of ASME N511-2007. For good, dry carbon the calculation is:

$$L = 100 \frac{C^{dn}}{C^{up}} \quad (\text{Eq. 1})$$

Where L is the % leak, C^{dn} is the downstream concentration, and C^{up} is the upstream concentration. There is no guidance in the current form of ASME N511 on the values to use for C^{dn} and C^{up} ; single point, average, or other. Typically for good, dry carbon in the absence of desorption they can be the average of all the C^{up} samples and the average of all the C^{dn} samples taken over the duration of the test. When there is breakthrough, using average values will cause the calculation of bypass leakage to *overestimate* the true result. A practical method to minimize this overestimation is to extrapolate the downstream concentrations back to time = 0 for a true measurement of bypass leakage. The value of time = 0 should be the time that the concentration upstream of the filter bank reaches equilibrium. The calculational method for strict averaging plus three alternative mathematical methods to best-fit the breakthrough curve are presented.

All of the calculations assume the criteria specified in ASME N509/N510/N511 and ASME AG-1-2012 are satisfied:

- Immediately upstream of the filter bank, the concentration variation is limited to +/- 20% of the average. (ASME AG-1-2012 TA-4642 and TA-4743)
- The downstream sample point is acceptable when concentrations across the sample plane do not exceed +/- 20% of the average. (ASME AG-1-2012 TA-V-4200 (g))
- The maximum and minimum upstream concentrations differ by less than +/- 20% of the average (ANSI/ASME 510-1980 9.4.6)

ASME N510 simply states that “the adsorber leak rate is determined from the ratio of downstream to upstream concentration” (section 10.2), while ASME N511 states to record concentrations until sufficient data have been recorded (Appendix IV (f)) and then lists Eq. 1 (Appendix IV (h)). Additionally, ASME N511 does not include subtraction of background concentrations in its current form as can be seen in equation 1. Strict reading of ASME N510 and N511 may lead one to believe that Method 1 – Average is the preferred calculational method. Method 1 assumes no breakthrough; the calculations are outlined in equations 2-5 below.

1. Method 1 - Average

$$L = 100 \frac{C_{avg}^{dn} - C_{bg}^{dn}}{C_{avg}^{up} - C_{bg}^{up}} \quad (\text{Eq. 2})$$

$$C_{avg}^{dn} = \frac{1}{n} \sum_{j=1}^n C_j^{dn} \quad (\text{Eq. 3})$$

$$C_{avg}^{up} = \frac{1}{n} \sum_{j=1}^n C_j^{up} \quad (\text{Eq. 3})$$

$$C_{bg}^{dn} = \frac{1}{nb} \sum_{j=1}^{nb} C_{bg_j}^{dn} \quad (\text{Eq. 4})$$

$$C_{bg}^{up} = \frac{1}{nb} \sum_{j=1}^{nb} C_{bg_j}^{up} \quad (\text{Eq. 5})$$

Where the subscripts *avg* and *bg* represent average and background, respectively while n and ng represent the number of test samples and the number of background samples, respectively.

Proper application of Methods 2, 3 and 4 to best-fit the breakthrough curve require higher quality data than required by ASME N510-511. Fitting breakthrough curves requires more stringent qualifications of the upstream and downstream injection and sample points/manifolds with regards to mixing, timing, and precision of measurement. In particular, the time for the upstream concentration to come to a uniform equilibrium across the filter bank is approximately one minute after the injection and the sampling program are simultaneously initiated. Additionally, the post injection upstream concentration should drop to 10% of the upstream equilibrium challenge concentration in approximately one minute after the injection is turned off.

A good indication that breakthrough is occurring is that the post downstream concentration stays elevated after the injection is turned off or may drop immediately but then starts to increase again. For this, one should take approximately 5 post readings. Coincidentally, the drop in the downstream concentration after the injection is turned off divided by the last upstream concentration before injection is turned off can also be used to calculate the leakage but again this is not supported by N510-N511 and is not described further here.

If breakthrough occurs, one of 3 empirical equations can be used to calculate the downstream concentration when the upstream concentration reaches equilibrium at the filter bank. These methods assume that the tracer concentration comes to equilibrium much faster than the rate of increase of downstream concentration due to breakthrough.

If Δt is the time after the tracer arrives at the filter bank and C_{lk}^{dn} is the downstream concentration immediately after the arrival of the tracer, then the following 3 equations can be selected to predict C_{lk}^{dn} using least squares regression.

2. Method 2 - Exponential Breakthrough

$$C(\Delta t) = C_{lk}^{dn} \times e^{\alpha \Delta t} \quad (\text{Eq. 7})$$

where:

$$\Delta t = t - t_{inj}$$

and t_{inj} is the time the upstream tracer reaches equilibrium at the filter bank

3. Method 3 - Square of Time Breakthrough

$$C(\Delta t) = C_{lk}^{dn} \times e^{\alpha \Delta t^2} \quad (\text{Eq. 8})$$

4. Method 4 - Quadratic of Time Breakthrough

$$C(\Delta t) = C_{lk}^{dn} \times e^{(\alpha \Delta t + \beta \Delta t^2)} \quad (\text{Eq. 9})$$

Using equation 2, the leakage is given by

$$L = 100 \frac{C(0) - C_{bg}^{dn}}{C_{avg}^{up} - C_{bg}^{up}} \quad (\text{Eq. 10})$$

Where $C(0) = C_{lk}^{dn}$, is calculated from either Eq. 7, Eq. 8, or Eq. 9 by regression analysis. A rigorous application of these procedures requires a statistical error analysis of the coefficients. R-squared (R^2) is a statistical measure of how close the data are to the fitted regression line. Results that have R^2 less than approximately 98% should be used with caution as the difference

between the predicted value and the true value may be too large. The current standards cited above provide no error analysis and no confidence limits of the results of the test data.

The leakage values for all the comparison filter tests in this paper were calculated using Method 2 – Exponential Breakthrough. The exponential curves provided the best fit for the breakthrough curves and the calculation works well whether or not there is breakthrough. To provide an example of the use and limitations of exponential breakthrough a plot for three tracers (PMCP, PMCH, and Vertrel) is provided in Figure 5 below. Note that the data is presented as % leak for each pair of samples, not C^{dn} concentrations, similar to Figures 2 and 3. Additionally note that the R^2 value, a measurement of how good the data fits the curve, shows excellent agreement for Vertrel but not for PMCP or PMCH. This is due to the fact that the breakthrough of PMCP and PMCH was too large to accurately calculate the leakage. However, for PMCH, if only the first three data points are utilized a reasonable breakthrough curve ($R^2 = 0.9758$) can be fitted with a resulting leakage calculation far closer to the true value.

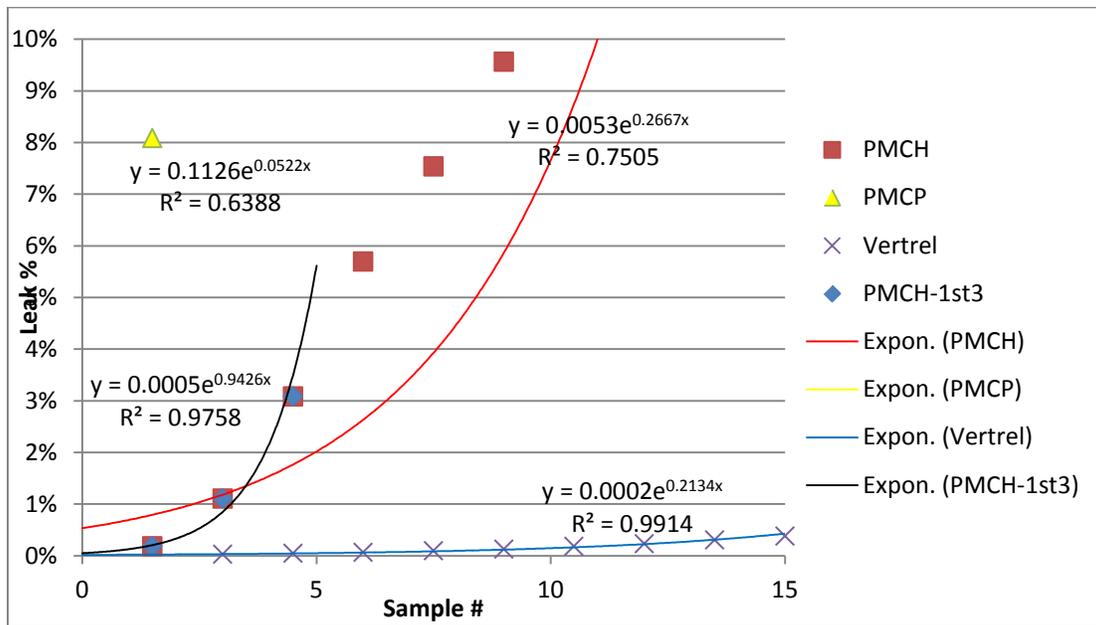


Figure 5 – Filter comparison test performed on 12/22/14, moisture loading = 24.5% by weight. PMCP calculated leak = 11% (overestimation), PMCH calculated leak = 0.51% (overestimation), Vertrel = 0.02% with $R^2 = 0.9914$. PMCH calculated leak using only the 1st 3 data sets = 0.05% with $R^2 = 0.9758$.

Figures 6 and 7 provided below are actual data outputs from the analysis performed by the LMP-3000 for the PMCH and Vertrel comparison tests performed on 12/22/14. Circles are used for the upstream concentrations, triangles for the downstream concentrations. The color black denotes background data, red the test data, and green the post data. The lines indicate the curves used for the fitting the data. The upstream dashed lines are the average of each data set. The downstream solid lines are a plot of the results of the model used to calculate the line. Note that due to heavy breakthrough for PMCH only the first three downstream data points were used in the calculation of $C(0)$.

As previously mentioned, using the average method will cause an overestimation in the calculation of actual leakage in a carbon AFU. Using the Vertrel data from the filter comparison test provided above, the average concentration method returned a leakage value of 0.15% vs. 0.02% for the exponential method. To show this graphically, the data for only the Vertrel series above is provided in Figure 8 below using the average and the exponential methods.

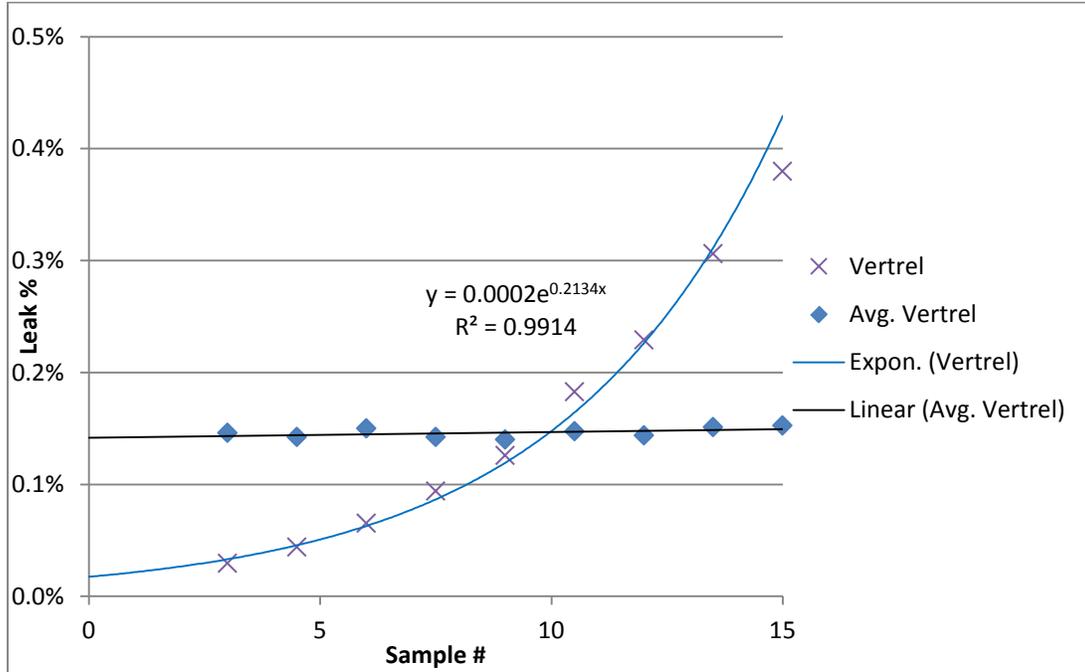


Figure 8 – Comparison of average vs. exponential calculation of leakage when breakthrough occurs. Test on 12/22/14, moisture loading = 24.5%, only Vertrel-XF data presented.

SUMMARY

All of the tracers in this study; PMCP, PMCH, Vertrel-XF, and PDCH satisfy the six selection criteria outlined in ASME AG-1 for alternative tracers. Additionally it was shown that certain higher MW and BP tracers are better retained on the carbon for performing leakage tests on AFUs with older or more moisture laden carbon. For testing AFUs where breakthrough is likely to occur the selection of a higher MW and BP tracer, such as PDCH, over R-11 will vastly improve the chances of performing a successful in-place filter test. Finally it was shown that using alternative calculation methods for cases with moderate breakthrough, such as the previously described Method 2, will greatly improve the accuracy of the reported leakage rate in comparison to a simple average calculational method which will likely overestimate the reported leakage. However, more stringent restrictions on the quality of the data are required to properly apply these alternative calculation methods.

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