

This article was downloaded by:

On: 25 January 2011

Access details: *Access Details: Free Access*

Publisher *Taylor & Francis*

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Separation Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713708471>

The Use of Simplified Statistical Processing Techniques to Increase Sensitivity of Detection to Particulate Breakthrough

Michael Sadar^a

^a Hach Company, Loveland, CO, USA

To cite this Article Sadar, Michael(2008) 'The Use of Simplified Statistical Processing Techniques to Increase Sensitivity of Detection to Particulate Breakthrough', *Separation Science and Technology*, 43: 7, 1713 — 1724

To link to this Article: DOI: 10.1080/01496390801973748

URL: <http://dx.doi.org/10.1080/01496390801973748>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

The Use of Simplified Statistical Processing Techniques to Increase Sensitivity of Detection to Particulate Breakthrough

Michael Sadar

Hach Company, Loveland, CO, USA

Abstract: The monitoring of filtration performance is often accomplished through the monitoring of the filtrate (effluent) stream for particulate materials that would be present in a compromised filtration system. Two technologies that are used to monitor for particulates are laser nephelometers and particle counters. These two particulate detection technologies have proven to be effective on certain types of filtration systems, including membranes. However, particulates that would travel through a compromised filtration system can be diluted to below detection levels. This paper provides a means of analyzing the raw laser turbidity signals using simplified statistical procedures that can help to regain sensitivity to the presence of particles under such conditions. The method was demonstrated to be effective in several different filtration integrity studies. Such statistical processing techniques are easily applied to existing instruments through algorithms that ultimately provide an additional means for detection of filtration breakthrough.

Keywords: Laser turbidity, breakthrough detection

INTRODUCTION

The use of particulate detection techniques is a key method for monitoring filtration effectiveness and detecting filtration breakthrough. Of the available techniques, those that focus on using light scatter from particles that would otherwise pass through a breached filter include turbidity and

Received 3 September 2007, Accepted 15 November 2007

Address correspondence to Michael Sadar, Hach Company, 5600 Lindbergh Drive, Loveland, CO 80539, USA. E-mail: msadar@hach.com

particle counting. Both can be applied as on-line (process) monitoring techniques. Within the past decade, newer laser-based techniques in turbidity analysis have emerged and provide more sensitive methods for monitoring filter performance. These laser-based technologies are able to identify filtration integrity problems earlier and with better detection levels.

The data that is produced using laser nephelometric turbidity can have a specific statistical analysis applied to it in real time to further enhance sensitivity to the detection of filter integrity loss.

BACKGROUND

Turbidity has been recognized as a simple and basic indicator of water quality. It has been used for monitoring drinking water, including that produced by filtration for decades. Turbidity measurement involves the use of a light beam, with defined characteristics, to determine the semi-quantative presence of particulate material present in the water or other fluid sample. The light beam is referred to as the incident light beam. The material in the water causes the incident light beam to be scattered and this scattered light is detected and quantified relative to a traceable calibration standard material. The higher the quantity of the particulate material contained in a sample, the greater the scattering of the incident light beam and the higher the resulting turbidity. Laser turbidimeters with high turbidity detection sensitivity can be effective monitoring tools for filtration integrity involving minute breakthrough events, but can become questionable with respect to the reliability to the prediction of a breakthrough event. Other more sensitive instrument technologies are available, such as particle counters based on light scatter, but the economics related to obtaining and applying those technologies often limits their use (1).

Any particle within a sample that passes through a defined incident light source (often a incandescent lamp, light emitting diode (LED) or laser diode), can contribute to the overall turbidity in the sample. The goal of filtration is to eliminate particles from a given process. When filtration systems are performing properly and monitored with a turbidimeter, the turbidity of the effluent will be characterized by a low and stable measurement. Turbidimeters which typically utilize incandescent or long wavelength light sources can be effective for the detection of particles down to a certain level of particle concentration, but they become less effective on super-clean waters, where particle sizes and particle count levels are very low. At these super clean levels of turbidity, the actual turbidity change that would result from a filter breach can be so small that it becomes indistinguishable from the turbidity baseline noise of the instrument.

This baseline noise has several sources including: the inherent instrument noise (electronic noise), instrument stray light, sample noise, and noise in the light source itself. These interferences are additive and they become the

primary source of false positive turbidity responses and can adversely impact the instrument detection limit.

The advent of laser nephelometry better addresses the need for low-level turbidity analysis in cleaner water samples. Laser turbidimeters (also known as laser nephelometers) possess enhanced optical designs that yield greater sensitivity and baseline stability. The primary distinction between a laser nephelometer and a basic nephelometer is found in the incident light source and the detector. The laser turbidimeter utilizes a highly collimated, laser-based light source that is primarily monochromatic. The characteristics of this light source allow the light energy to be concentrated and focused into a very small volume within the sample chamber in a given instrument. This combination provides an incident beam with a high power density, which is efficiently scattered by particles within a sample. The detector is also of greater sensitivity and provides greater response to scattered light. Preferably, the peak of the detector response spectrum should completely overlap the spectrum emitted by the incident light source to generate maximum optical sensitivity. This combination of detector sensitivity, collimated light source, and the high power density of the incident light source provides for a very high signal-to-noise ratio for the laser turbidimeter. This signal-to-noise ratio enhances the sensitivity to detect very small changes in turbidity that can be distinguished from a very stable measurement baseline (2). Thus, if the baseline variability is minimal with respect to instrument noise, such variability on clean particle-free water will also be minimal.

Laser turbidimeters, and other instruments that provide high signal-to-noise ratios, will yield extremely stable measurement baseline levels in comparison to traditional turbidimeters. Stable baselines allow for the detection of very fine changes in the turbidity within a sample that would otherwise be indistinguishable with conventional turbidimeters. Further, this baseline can be characterized in terms of stability and then serve as an additional analysis parameter. This parameter would complement the directional trending of the turbidity measurement value itself.

Technical literature from a leading turbidimeter manufacturer who has commercialized both laser and traditional turbidimeters provides confirmation of the increased sensitivity of laser turbidimeters through limit of detection (LOD) specifications. The limit of detection is defined as the smallest change that an instrument can detect that is due to the analyte, which is in this case turbidity (3). In this comparison, the turbidimeter with a traditional design has a stated LOD of 3.2 milli-nephelometric turbidity units (mNTU) and the laser turbidimeter design has a stated LOD of 0.3 mNTU (4, 5). An American Water Works Association Research Foundation Study evaluated different membrane integrity monitoring techniques which included laser and traditional turbidimeters. The study provided evidence that a laser turbidimeter was able to better detect membrane breaches than traditional turbidimeters (6). It was not until

the development of laser-based instruments that were capable of producing extremely stable measurement baselines that the variability of the measurement itself could be studied from a quantitative and qualitative aspect. This variability can be demonstrated to aid in the prediction of filter breakthrough events.

MONITORING TECHNIQUES FOR FILTRATION BREAKTHROUGH

When measurements are performed in a process setting, several methods can be used to analyze and interpret the data. These are

1. Monitoring the measured value for a distinct step change;
2. Monitoring the trend in the measured value over a distinct increment of time; and
3. Comparing an interval of data against a pre-established baseline.

These approaches are used to evaluate the filtration process and often present a responsive action based on the logged information. This presents the concern that the reaction time to an "event" may be too slow.

A novel approach to real-time data analysis has been developed that uses basic statistical processing techniques to help predict the impending change in filtration performance prior to its gross failure. This technique processes and analyzes the variability of the laser turbidity signal itself and treats it as an independent parameter. This is then correlated to filtration performance in terms of the detection and qualification of any filter breakthrough event.

When monitoring a sample as it leaves an intact filtration system, its laser turbidity baseline should be characterized as quiet and stable. When a filtration breakthrough occurs, this turbidity or particle count level (i.e. counts) should increase as the inflow of particles pass through the failed filtration boundary. However, as the breakthrough event begins, only a few particles will trickle across the filtration boundary. These few particles are not sufficient to increase the raw turbidity or particle count reading above its established baseline, but they are sufficient to cause the baseline to show unsuspected variability.

Three experiments are presented in this paper that help to demonstrate the use and application of the variability of a laser turbidity baseline for use in the detection of filtration breakthrough. One experiment evaluated the filtration run of an anthracite filtration system for drinking water applications. The second experiment involved the use of latex-based particles to determine sensitivity of different algorithms that assess the severity of the variability in the laser turbidity measurement. The third experiment involved full-scale membrane integrity testing.

MATERIALS AND METHODS

Methodology for the Measurement and Calculation of Baseline Variability

The technique for enhancing the detection of a pending filtration breakthrough is a simplified process statistical model. This model has successfully been applied to both process laser turbidity or process particle counter applications (but focus here will be primarily on laser turbidity). The baseline variability is simply quantified and treated as a separate monitoring parameter. The baseline variability parameter can also provide qualitative information regarding the nature of the filtration breach. The variability is sensitive to both particle size and count and thus provides information regarding the material that is passing through a breached filter.

The measurement of variability is quantified using a simple statistical process parameter known as the percent relative standard deviation or RSD. The RSD is calculated as the standard deviation for a given set of measurements divided by the average for the same set of measurements. The quotient can then be multiplied by 100 to express this result as a percent. See equation (1) below:

$$\text{RSD} = (\text{Stdev}_n / \text{Av}_n) \times 100 \quad (1)$$

Where n = a defined number of measurements that are used to calculate both the average and the standard deviation.

The RSD parameter, when applied as a process measurement parameter is subject to several variables that can impact its relative sensitivity. These include

1. the number of measurements taken each time to generate the RSD value;
2. data set filtration;
3. data overlap;
4. measurement frequency; and
5. data logging rate.

Number of measurements: the number of measurements used to generate the RSD value will impact its sensitivity and response time. If the RSD value is generated using a very small data set (2–4 values), the resultant baseline will contain a significant amount of inherent noise. This noise can mask the sensitivity in much the same way that a poor signal to noise ratio impacts the sensitivity of a process turbidity measurement. If too many values (15–20 values) are used to generate the RSD measurement, then the response time to an impending particle event can be delayed.

Data set filtration: depending on the application of the analytical measurement, the sample may be inherently noisy and require pre-filtration of the data prior to performing the RSD calculation. For example, a sample that

contains a significant amount of bubble interference may have such a high level of noise in the baseline that the variability may be lost. In such a case, a large number of measurements may be taken and any value above a pre-set level is excluded from the data. The remaining values are then used to generate the RSD value. In turbidity measurement electronic bubble reject algorithms that exclude singular random spikes are often used which is an example of pre-filtration of data.

Measurement frequency: data filtration applications will be more successful on technologies with high measurement frequency. The frequency of measurements will impact the performance of the RSD parameter because data is often lost under slow (defined as 1/minute or slower) measurement frequencies. For example, if the measurement frequency is very slow, then filtration of this data may defeat the purpose of use for detection of breakthrough. However, if the frequency rate is excessively fast, it may incorporate much of the systematic noise and an excessive rate of filtration may be required. The choice of measurement frequency is typically dictated by the application.

Full-Scale Filtration Monitoring for Breakthrough

In this experiment, full-scale anthracite-based filtration monitoring was conducted using traditional process turbidimeters, a laser turbidimeter, and a light-obscuration particle counter. The filtration mechanism used was a dual-media filter technique in which 12 inches of sand are overlaid with 36 inches of anthracite coal. The anthracite itself is further distributed into layers based on particle size with the smallest particles near the bottom and the largest particles at the top of the filter. This filter, when aided by chemical pre-treatment is efficient at removing particles greater than 1 μm . All measurements for turbidity and particle counting were logged every minute throughout an entire filter run (backwash to backwash). Measurements from each instrument were recorded at the same interval of one minute. The analysis of this data is presented in the results section.

Laboratory Experiment with Polystyrene Latex Spheres

This experiment involved the controlled injection of polystyrene latex (PSL) microspheres of a mean diameter of 1.03- μm into a stream of reverse osmosis-filtered water. The microsphere concentration spike was known, as was the flow rate of the water that was spiked. Thus the final concentration of microspheres was calculated and was held constant at 40 counts per mL. The spike was initiated after turbidity and RSD baselines were first established. After the water was spiked, it flowed through a manifold that split into three equal flow streams that led separately to three laser turbidimeters at the rate of 150 ml/minute to each laser turbidimeter. Each laser turbidimeter had used a

different algorithm to generate the RSD. This data was then evaluated to determine which algorithm produced the best true positive result with the best sensitivity.

Full-Scale Membrane Integrity Monitoring

The monitoring of membrane systems for filtration integrity is necessary and required in the production of water for human consumption. The determination of detection levels for either laser turbidimeters or particle counters is critical for such applications. The most common method for determining instrumental detection of membrane filtration failures is through fiber cutting/pinning (referred to as fiber cutting) tests.

The test membrane system contained two modules. Each module contained ultra filtration fibers with a nominal pore size that was less than 0.05- μm . One module had no severed fibers. The other module contained several pre-cut membrane fibers. The two modules were incorporated into a common housing. Combined, the modules contained approximately 40,000 fibers that were available for filtration and produced approximately 50 gallons of filter effluent per minute. The effluent was monitored by a laser turbidimeter that was equipped with an algorithm to record both the laser turbidity value and the RSD response. The values were recorded at one-minute intervals.

Fiber cutting involved the deliberate severing of a known number of fibers within a membrane module. The damaged membrane was then brought back on-line and the filtrate was monitored using the test instrumentation to establish a baseline of lost integrity. The module was then repaired in a series of steps; usually one fiber at a time, and a new filtrate baseline was re-established after each repair. This process continued until the integrity of the module was completely restored. It was expected that eventually a level of repair would be reached in which the filtrate monitoring with laser turbidity would fail to change. It is at that level of integrity that the instrument limit of detection (LOD) was determined.

DATA AND RESULTS

Filtration Monitoring for Breakthrough

In Fig. 1, the turbidity and particle count levels of the effluent water that are exiting an anthracite filter are monitored over time. As the filter run progresses, increases in the turbidity and particle count levels were observed but these changes were not indicative of any pending breakthrough (7). However, if the focus is drawn to the behavior of the individual baselines, the variability of each parameter increases as the run progresses. This baseline variability is often discarded as inherent instrument noise, when in reality it is a reflection of the filtration process.

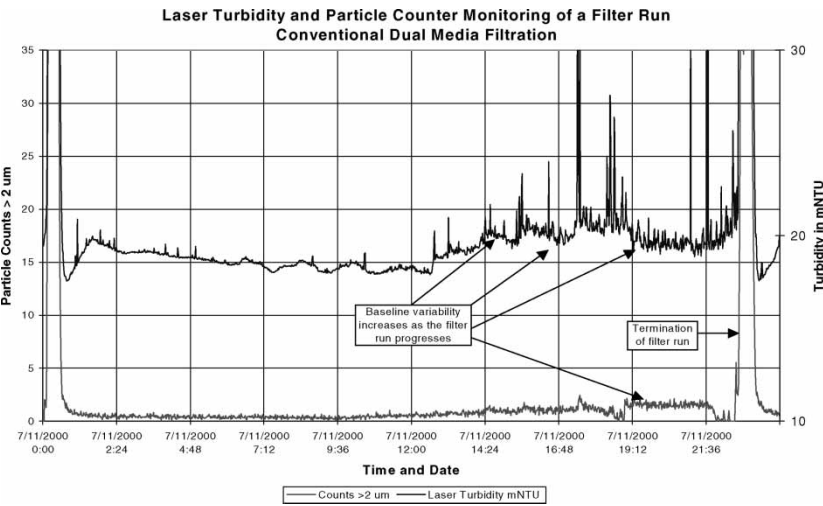


Figure 1. Turbidity and particle count monitoring of a typical filter run from an anthracite-based multi-media filter. This is a common filtration technique for drinking water plants.

Laboratory Experiment with Polystyrene Latex Spheres

Figure 2 provides an example showing data can be treated to deliver different responses. In this figure the left y-axis represents laser turbidity response and the right y-axis represents the RSD response. Time is presented on the x-axis

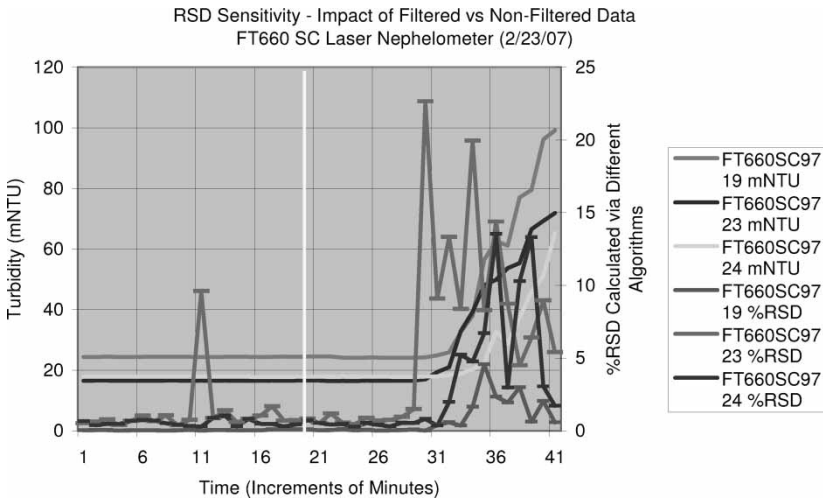


Figure 2. The comparability in response by different RSD algorithms to a particle spike.

and is in increments of minutes. The time when this spike was initiated was at $t = 20$ and a yellow horizontal line represents the starting point.

Laser turbidimeter 9719 used an RSD algorithm that was generated using data that was pre-filtered to remove false positive spikes that would be caused from bubbles. This algorithm displayed minimal response to the spike of 1.03- μm particles. Laser turbidimeter 9723 used an algorithm that was generated from ten consecutive data points. This algorithm was oversensitive and generated false positive spikes prior to the time the injection started. However, the algorithm did respond first and prior to any changes in the actual turbidity. Laser turbidimeter 9724 used an algorithm that was generated from ten consecutive data points. After the collection of the 10 points, the top two and bottom points were eliminated and the RSD was generated from the remaining seven points. This algorithm also responds before the turbidity parameter responds to the spike, but it does not generate false positives.

This experiment demonstrated that the RSD algorithm could be made more or less sensitive, depending on the treatment of the turbidity data. This experiment also demonstrated that the RSD parameter can show a response prior to the turbidity baseline itself which can buy valuable time when responding to an impending filtration breach. The instrument known as the FilterTrakTM 660 laser nephelometer and incorporated the algorithm that responded best to the spike of particles that was conducted in this study.

Membrane Integrity Monitoring

Figure 3 displays the data that was derived from the membrane integrity experiment. For each level of integrity, the laser turbidity and RSD for the filtrate sample stream was plotted. The left-hand axis of Fig. 3 displays the laser turbidity (laser turbidity in mNTU) and is represented by the red trace. The right-hand axis displays the RSD response, which is represented by the green trace.

The data in Fig. 3 represents the steps for pinning each of the severed fibers in chronological order from left to right. Black vertical dashed lines separate each test. The upper control limits (UCL) for each pinning parameter were calculated and plotted on the graphs. These are represented by the thicker green and red horizontal traces.

The UCL values were also derived from the baseline data that was established under an integral filtration system that was void of any breaches or defects. For the specific parameter, the average and standard deviation was calculated for the integral baseline run. Each standard deviation was multiplied by three and added to the averaged value (for the same parameter) for this same baseline. Statistically, this generated upper confidence limit of 99 percent. Equation (2) summarizes this UCL calculation (8).

$$\text{UCL} = \text{Average}_{\text{run}} + 3 * \text{Std Deviation}_{\text{run}} \quad (2)$$

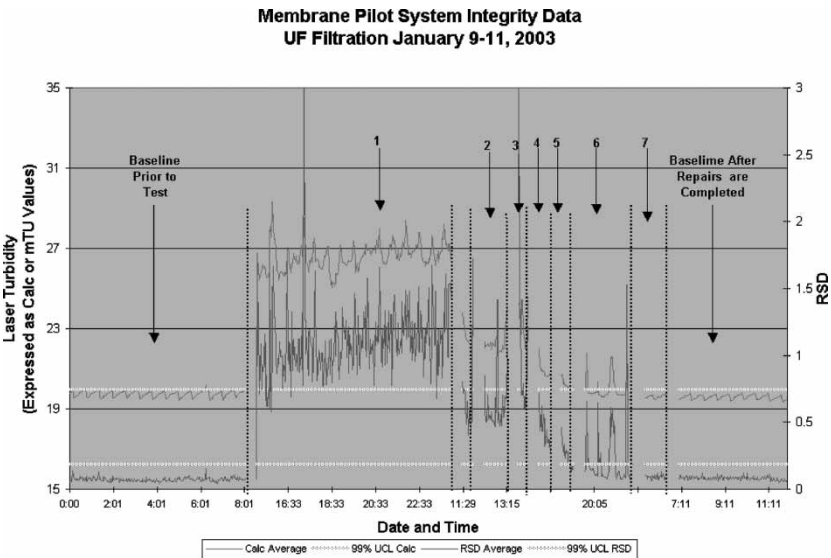


Figure 3. Graphical display of laser turbidity and baseline variability monitoring for membrane integrity loss during a series of fiber cutting tests. Numbered Events are as follows from left to right: Event 1—more than four cut fibers plus two pinholes; Event 2—exactly four cut fibers plus two pinholes; Event 3—three cut fibers plus two pinholes; Event 4—two cut fibers plus two pinholes; Event 5—one cut fiber plus two pinholes; Event 6—two pinholes only; Event 7—one pinhole only.

In this study, the baseline condition in which the membrane was integral was the basis for determining the UCL for both the laser turbidity and the RSD parameters. To calculate this value for the respective parameter, the entire data set for each was used to determine the average and standard deviation. The standard deviation was then multiplied by a factor of three and added to the average value. The result generated the UCL. A factor of three was selected as the criteria for the experiment because it approaches a 99 percent confidence limit for a single tail test, which was the pre-determined requirement for this study. Each UCL was considered to be the threshold for the detection limit for the respective RSD and laser turbidimeter parameter. If the response to a given integrity test exceeded the UCL limit, then the response was sensitive at least to that level of membrane integrity. The UCL is also referred to as the limit of detection (LOD) for this system.

In this study, the RSD and turbidity LODs were exceeded at all levels of cut fibers, denoted in Fig. 3 as events 1 through 5. The two-pinhole breach, represented by event 6, also resulted in a positive detection for parameters. Figure 3 illustrates the complementary nature of these two parameters, which provides confidence in the detection capabilities of this technology.

Table 1 displays the averaged relative response for the laser turbidity and the RSD parameter as a function of the number of severed membrane fibers.

Table 1. Calculated average response for each integrity test performed on the UF module

Number of cut fibers ^a	Percent cut fibers	Turbidity: relative change over baseline (LOD = 1.85%) ^b	RSD: relative change over baseline (LOD = 106%) ^c
0.1	0.00001%	1.85%	110.02%
0.2	0.00001%	3.01%	183.95%
1.2	0.00006%	6.46%	426.31%
2.2	0.00011%	18.70%	1184.61%
4.2	0.00021%	13.89%	671.11%
5.2	0.00026%	35.64%	1218.51%

^aA pinhole in a fiber was equated to 0.1 cut fibers.
^bThe turbidity UCL of 1.85% is the minimum positive increase in the baseline necessary to confirm a positive response to the integrity loss.
^cThe RSD UCL of 106% is the minimum positive increase in the baseline necessary to confirm a positive response in the integrity loss.

For a given number of severed fibers, the response of the RSD parameter was between one and two orders of magnitude greater than the turbidity response. The additional sensitivity becomes significant when the filtrate from several membrane modules are combined and dilution of particles that would freely pass through a single breach takes place.

CONCLUSIONS

The advent of Laser turbidimeters and have improved the detection of loss in filtration integrity. These instruments possess highly improved optical qualities from traditional nephelometry to produce a very stable process measurement system. This enhanced stability provides additional information that can be deciphered from the laser turbidity measurement itself and used as a separate parameter to further improve the limit of detection to breakthroughs in filtration systems. This parameter is known as the RSD parameter. The parameter has been shown to also enhance the sensitivity of detection of minor breakthroughs in different filtration systems. Studies in conventional anthracite filtration, micro-filtration, ultra-filtration, nano-filtration, and reverse-osmosis filtration have proven out this process detection parameter.

The ability to apply this RSD parameter as a predictive indicator of pending filtration breakthrough leads back to the optical view volume that is generated inside the analysis sensor. The small and very defined view volume is creates a high energy density of incident light bean. This beam is capable of detecting the presence of particles in low concentrations. The RSD parameter is primarily sensitive to particles that have a diameter

greater than 1.0- μm , but the laser turbidity signal is sensitive to particles as small as 0.01- μm . Combined the two parameters can see very small breaches in filters, such as pinholes.

Laser turbidimeters are designed to meet these criteria and along with particle counters, can use the real time derivative of their monitoring baselines as an independent indicator of a membrane breach. One commercially available laser turbidimeter offers RSD parameter, which is the Hach FilterTrakTM Laser nephelometer.

ACKNOWLEDGMENTS

A significant effort was made by many individuals to develop and test the applications of the various technologies associated with this study. This included the chemists, engineers, technical writers, and technical illustrators at Hach Company who helped conduct the testing, review data, and present the results. Special thanks also goes out to the Leah Oxenford of Hach Company for her efforts in generating many of the results that were presented. In addition Mark Lichwardt of Burns & McDonnell and Frank Watt of the City of Thornton, Colorado's Columbine Water Treatment Facility for providing the opportunity to test the methodologies presented in this paper on several different membrane pilot plants.

REFERENCES

1. Clesceri, S., Greenberg, L., and Eater, A. (1998) *Method 2130B. Turbidity, Standard Methods for the Examination of Water and Wastewater*, 20th Edn.; American Public Health Association: Washington, DC.
2. Sadar, M. (1999) *Introduction to Laser Nephelometry; An Alternative to Conventional Particulate Analysis Methods*; Loveland, CO. Lit. No. 7044.
3. International Standardization Organization. (2006) *Method 15839:2003, IDT Water Quality—On-line Sensors/Analyzing Equipment for Water—Specifications and Tests*; International Standardization Organization: Brussels.
4. Hach Company. (2006) *1720E Low Range Turbidimeter User Manual*; Hach Company: Loveland, CO.
5. Hach Company. (2005) *FilterTrakTM660sc User Manual*; Hach Company: Loveland, CO.
6. Sethi, S. et al. (2004) *Assessment and Development of Low-Pressure Membrane Integrity Monitoring Tools*; American Water Works Association Research Foundation: Denver, CO.
7. Sadar, M. (2000) A Strategy for Optimizing WTP Performance Using New Light Scatter Technologies, Proceedings from the 2000 Water Quality Technology Conference, Salt Lake City, UT.
8. Sadar, M. and Harrington, L. (2002) Evaluation of a New Continuous On-Line Integrity Testing Method for Membrane Filtration, Proceedings from the Water Quality Technical Conference, Seattle, Washington.