

Intercomparison of Ambient Optical Monitoring Techniques

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INTRODUCTION

A major concern in the field of visual air quality monitoring is the ability to make a truly unmodified, continuous measurement of the ambient atmospheric extinction (bext), scattering (bscat), and absorption (babs) coefficients. Optical instrumentation is, for the first time, approaching this goal. A unique field study was held at Shenandoah National Park during the summer of 1991 to intensively examine eastern regional haze and acid aerosols. A major focus of the study was an investigation of the role liquid water bound to ambient aerosols plays in visual air quality degradation. To help in this assessment, an intercomparison of current and newly developed optical monitoring techniques was conducted. Instrumentation tested included:

- Three transmissometers: two operating over a 0.67 km path, and one over a 1.41 km path;
- A Belfort 1590 nephelometer, operating with a dual switchable inlet: heated and ambient;
- A newly-developed OPTEC ambient nephelometer; and
- Three relative humidity measuring sensors: Campbell 207, Rotronics MP-100MF and an aspirated wet/dry bulb system.

This paper will present detailed descriptions of the instrumentation and data collection protocols followed. Data analyses will include intercomparison of measured optical parameters with emphasis on the performance, uncertainties, and limitations of each monitoring technique. The analyses will incorporate data collected from the 1991 summer intensive, historical data from national visibility monitoring programs, and laboratory investigations into the precision and accuracy of the techniques.

INSTRUMENTATION

Transmissometers

Transmissometers directly measure the irradiance of a light source after the light has traveled over a finite atmospheric path. The transmittance of the path is calculated by dividing the measured irradiance with the calibrated initial intensity of the light source. The average extinction of the path is calculated from the transmittance and length of the path. It is attributed to the average concentration of atmospheric gases and ambient aerosols along the path.

Transmissometers make a completely ambient measurement of bext without perturbing or selectively sampling the atmospheric aerosol. However, transmissometers require path lengths of a few kilometers to achieve the sensitivity to resolve extinctions near the Rayleigh limit. In areas with non-uniform distribution of aerosols, comparisons of measured extinction and reconstructed extinction from concurrent particulate samples can often be misleading. Extinction measurements from transmissometers also are effected by any meteorological or optical interferences present along the path which are independent from the ambient aerosol.

The OPTEC, Inc., LPV-2 long-path transmissometer has been in use since 1986.^{1,2,3,4,5} There are currently over thirty instruments operating in various visual air quality monitoring programs in North America, from highly polluted urban areas to pristine wilderness environments. Three transmissometers were used in the summer 1991 intensive study; two monitored the extinction along side-by-side 0.67 km paths, the third operated over a parallel 1.41 km path offset approximately 100 m from the short path.

Integrating Nephelometers

Integrating nephelometers estimate bscat by directly measuring the light scattered by aerosols and gases in an enclosed sample volume. The scattered radiation is integrated over a large range of scattering angles in a defined band of visible wavelengths. Since the total light scattered out of a path is the same as the reduction of light along a path due to scattering, the integrating nephelometer gives a direct estimate of bscat.

The nephelometer was initially devised during World War II by R.G. Beuttell.⁶ The first commercially available instrument was developed during the 1960s by Ahlquist and Charlson,⁷ and has been marketed as the MRI/Belfort 1550, 1560, and 1590 series nephelometers, with the last upgrades in 1979. The integrating nephelometer is the only visibility measurement technique approved as a "standard method" by the TE-5 Visibility Committee of the Air and Waste Management Association.⁸

While nephelometers have been used extensively for nearly twenty years, a number of severe limitations compromise the scattering data they collect. The following are major deficiencies that have been documented by many researchers:^{9,10,11,12,13}

- Modification of the ambient aerosols by heating, especially at high relative humidity when a large fraction of the ambient aerosol is composed of hygroscopic or volatile particles.
- Inlet, sampling train, and optical chamber design can cause an ill-defined size cut to the ambient aerosols accepted into the optical chamber.

- An inherent design restriction of all integrating nephelometers that prevents the measurement of light scattered in the extreme forward and backward directions. This limitation, known as truncation error, has its most severe effect on the proper measurement of scattering due to particles larger than a few microns in diameter.
- The wavelength response of the instruments' lamps, filters, detectors, and electronics have at times been poorly defined, introducing large uncertainties into scattering intercomparisons.
- Outdated electronics that display large non-linear drifts in zero and span values when operating the systems at ambient temperatures dramatically increasing the uncertainty of measured bscat.

Belfort Integrating Nephelometer: In an attempt to minimize the problem of inadvertent heating of ambient aerosols that has been associated with the use of Belfort nephelometers, modifications to the instrument, shelter, sampling train, and operating procedures have been developed to run the system as close to ambient as possible.¹⁴ The instrument is mounted in a ventilated shelter (Figure 1). The interior walls of the shelter are insulated, and the exterior coated with highly-reflective, white paint to minimize heating by solar radiation. A high-velocity ventilation fan continuously moves filtered ambient air through the shelter. This maintains the interior of the shelter and the exterior of the nephelometer and sampling train at the outside ambient temperature. In addition, the main nephelometer assembly must also be ventilated continuously with a separate, small exhaust fan. This removes heat generated by the lamp from the area inside the nephelometer that surrounds the optical chamber. The sampling train is kept as short as practical, and the portion of the inlet outside of the shelter is wrapped with reflective mylar tape to keep solar radiation from heating the tubing and the ambient aerosol as it traverses the inlet. The optical chamber temperature is monitored with a thermocouple placed as close to the scattering volume as practical without affecting the bscat measurement. These modifications only address the problem of heating of the ambient aerosol and none of the other concerns listed above. In fact, attempting to operate Belfort nephelometers near ambient conditions exacerbates the increased measurement uncertainty associated with outdated, unstable electronics.^{13,14}

Table 1 lists the mean chamber-inlet temperature differences that have been achieved by these systems in various monitoring programs during the past five years. Even with the above expensive, power-consuming modifications, the ambient aerosol is subjected to approximately 2° to 3° C heating during warm weather operation, and about 1.0° C heating during cold weather operation. It is interesting to note that the four non-ambient systems that were used in the 1990 PREVENT study had mean heating of about 13° C. This is a greater heating of the ambient aerosol than was reached during this study with the use of an in-line heater!

During the 1991 summer intensive, a Belfort 1590 nephelometer was switched hourly between ambient operation (as described above) and a heated mode when the sampled air stream was routed through an in-line heater. The mean chamber-inlet temperature differences for the ambient and heated modes were 1.5° C and 5.5° C respectively.

OPTEC NGN-1 Ambient Integrating Nephelometer: The Optec NGN-1 ambient nephelometer has been developed to minimize modification of ambient aerosol and address the other problems associated with the Belfort nephelometers: sizing by the inlet, large truncation error, poorly defined optical response, and outdated, unstable electronics. The system incorporates sensors, electronics, signal modulation and detection techniques developed for the Optec transmissometer. It features 12VDC low-power operation, solid compact design, and digital electronics giving a very stable linear performance over a wide temperature range. The complete system is contained in a single unit (Figure 2). An environmentally sealed compartment contains the CMOS computer, lamp assembly, motors, pumps, and electronics. The optical chamber features a single large door that opens a complete side of the chamber to unrestricted ambient air flow. The internal computer controls all operating functions and outputs data and systems parameters in digital and analog format. The optical design of the detector field of view, illumination cone, and cylindrical scattering volume allows for integration of scattered light from 5 to 175 degrees.

Laboratory tests on the prototype NGN-1 that was used during the 1991 summer intensive have indicated mean chamber-inlet temperature differences of about 1.5° to 2.0° C. This value is similar to the ambient Belfort system that also operated during the study. The second generation Optec NGN-2 nephelometer incorporates modifications that lower this heating to less than 0.5° C. These modifications are:

- increased thermal insulation between electronic chamber and optical chamber;
- improved optical chamber geometry and increased flow rate for a more laminar air flow through the system;
- more efficient duty cycle for motors and pumps to lower heating of electronic chamber; and
- increased heat sink capacity to allow for greater heat transfer to exhaust flow.

TABLE 1
 BELFORT SERIES (1550-1590) AND OPTEC
 INTEGRATING NEPHELOMETER OPERATING CHARACTERISTICS

Monitoring Period	Study Location	Series	Type	Mean Delta T deg C	Number Calibrations	Estimated Precision
11/87 - 1/88	Denver Brown Cloud					
	Federal Center	1590	Ambient	0.3	16	22%
	17th St. Plaza	1550	Ambient	-	17	16%
	Welby	1550	Ambient	-	19	18%
	Auraria	1560	Ambient	-	14	18%
	BAO Tower	1590	Ambient	-	17	25%
	Brighton	1560	Ambient	-	12	53%
9/89 - 1/90	Phoenix Brown Cloud					
	Scottsdale	1560	Ambient	2.8	11	49%
	West Phoenix	1560	Ambient	1.9	8	47%
	Tucson	1590	Ambient	3.1	7	28%
	Valley Bank	1560	Ambient	-	13	50%
	ICA	1590	Ambient	2.3	16	34%
	ICA	1550	Heated Inlet	5.0	15	125%
6/90 - 9/90	PREVENT					
	Mt. Rainier	1590	Ambient	2.3	19	10%
	North Cascades	1590	Ambient	1.5	8	32%
	Carbon River	1590	Ambient	2.1	14	5%
	South Mt.	1590	Non-Ambient	13.2	26	13%
	Dog Mt.	1590	Non-Ambient	11.3	21	14%
	Paradise	1590	Non-Ambient	13.5	21	35%
	New Halem	1590	Non-Ambient	8.7	17	16%
3/89 - 3/91	South Lake Tahoe	1590	Ambient	1.2	19	28%
10/90 - 3/91	Bliss Park, CA	1590	Ambient	0.6	9	11%
6/91 - 9/91	Shenandoah NP	1590	Ambient	1.5	8	20%
		1590	Heated Inlet	5.5		
		OPTEC NGN-1	Ambient	1.5	17	11%
1/92 - 3/92	MOHAVE					
	Amboy	OPTEC NGN-1	Ambient	0.9	4	17%
	Joshua Tree	OPTEC NGN-2	Ambient	0.3	4	8%
	Cajon Pass	OPTEC NGN-2	Ambient	0.7	3	5%

Relative Humidity Sensors

The importance of the effect of relative humidity on the scattering properties of aerosols cannot be overstated.¹⁵ Accurate relative humidity measurements are mandatory for a proper understanding and comparison of ambient optical measurements. Recent advances in the design and manufacture of low-power thin film capacitive relative humidity sensors have provided the means to more easily obtain accurate measurements of relative humidity. Sensors of this type have historically been plagued by non-linear response, hysteresis, creep, and instability; particularly at high humidity levels.

Ambient temperature and relative humidity measurements were made with three systems during the Shenandoah study. The first was an old-style capacitive sensor--a Campbell 207. The second was a new model by Rotronics--MP-100MF. This sensor features temperature compensation and a new polymer engineered to minimize hysteresis and creep. The third system was an Assman model 5230 psychrometer modified for continuous, unattended operation. Modifications included a large water reservoir, type E fine-wire thermocouples affixed to each bulb, and a low-power ventilation fan. Wet and dry bulb temperatures were logged with a Campbell Scientific 21X micrologger equipped with an internal thermocouple reference junction.

DATA COLLECTION AND AVERAGING PROTOCOL

All the instrumentation was operated continuously during the intensive monitoring period. Data was collected as one-minute average values for each instrument and sensor. The two short-path transmissometers operated over identical 0.67 km paths. The long-path instrument operated over a 1.41 km path offset about 100 meters from the short path. The two nephelometers were located next to each other, and the other aerosol sampling equipment about 1.5 km from the transmissometer receivers. Transmissometer lamps were changed monthly. All nephelometers completed automatic 15 minute clean air tests every six hours. The Optec nephelometer was calibrated twice a week, the Belfort system less frequently. Scheduled maintenance was accomplished following existing standard operating procedures and protocols.^{14,16} Spare systems were maintained on site to minimize data loss due to equipment failure. The Belfort nephelometer operated in a switching mode, alternating hourly between ambient and heated inlets. The temperature of the Belfort optical chamber was monitored continuously. The chamber-ambient temperature difference (ΔT) was calculated for each minute. The one-minute data from this system were examined to determine the best averaging period to create hourly averaged values of b_{ext} , b_{scat} , and relative humidity. Figure 3 displays the mean one-minute values of b_{scat} and ΔT from the Belfort nephelometer for the ambient and heated cycles. It was determined that due to the lag in this system reaching a stable state, only data collected during the last thirty minutes of each hour from each instrument would be used to calculate hourly means and uncertainties. All data was examined and coded to identify periods of instrument malfunction, maintenance, clean air or span periods, or other interferences.

ESTIMATED PRECISION AND ACCURACY

An analysis of the various optical techniques must include an assessment of the precision and accuracy associated with each measurement. The following estimates have been developed using standard uncertainty analyses¹⁷ and experience gained from investigations into the operation of transmissometers and nephelometers in various laboratory and monitoring programs during the past five years.

Transmissometers

Operationally, the basic equation used to calculate path transmittance is:

$$T = I_r / (F_{lamp} * I_{cal}) \quad (1)$$

T = Transmittance of atmospheric path r
 I_r = Intensity of light measured at r
 I_{cal} = Calibration value of transmissometer
 F_{lamp} = Variability function of lamp output

The relative uncertainty (U_x) of any measured parameter X is defined as:

$$U_x = \sigma_x / \bar{x} \quad (2)$$

\bar{x} = arithmetic mean of all x measurements
 σ_x = precision of measurements of X

Using propagation of error analysis, the relative uncertainty of the path transmission can be calculated from the relative uncertainties of the measured variables as:

$$U_T = (U_{I_r}^2 + U_{I_{cal}}^2 + U_{lamp}^2)^{1/2} \quad (3)$$

U_T = relative uncertainty of T
 U_{I_r} = relative uncertainty of I_r
 $U_{I_{cal}}$ = relative uncertainty of I_{cal}
 U_{lamp} = relative uncertainty of F_{lamp}^{cal}

To understand the uncertainty of a transmittance measurement requires a thorough investigation of the precision of each of the following:

- precision in calibration to determine I_{cal} ;
- precision in the measurement of I_r ; and
- precision in the measurement of F_{lamp} .

Relative Uncertainty of I_{cal}: The precision in calibration value I_{cal} can be determined by investigating the calibration equation. I_{cal} is the value that would be measured by the transmissometer detector if the atmospheric path was a vacuum. I_{cal} incorporates the path distance r, transmission of all windows in the path, and size of working aperture used:

$$I_{cal} = (CP/WP)^2 \times (WG/CG)^2 \times (WA/CA)^2 \times WT \times (1/FT) \times (1/T) \times CR \quad (4)$$

Using propagation of error analysis, the relative uncertainty is:

$$U_{I_{cal}} = (2U_{CP}^2 + 2U_{WP}^2 + U_{WG}^2 + U_{CG}^2 + 2U_{WA}^2 + 2U_{CA}^2 + U_{WT}^2 + U_{FT}^2 + U_{CR}^2)^{1/2} \quad (5)$$

Path distances are measured using a laser range finder. Apertures are measured with a precision micrometer. Gain settings are measured with a precision voltmeter. Window and neutral density filter transmittance is measured with a reference transmissometer by differencing techniques; thus, they do not require absolute calibration. The standard deviation of the raw readings (CR) are calculated at each calibration. Typical working values, measurement precision, and relative uncertainties of these values are:

Parameter	Value	Precision	Relative Uncertainty
CP Calibration Path	0.3 km	1 x 10 ⁻⁶ km	3.3 x 10 ⁻⁶
WP Working Path	5.0 km	1 x 10 ⁻⁶ km	2.0 x 10 ⁻⁷
CG Calibration Gain	100	1 x 10 ⁻²	1.0 x 10 ⁻⁴
WG Working Gain	500	1 x 10 ⁻²	2.0 x 10 ⁻⁵
CA Calibration Aperture	100 mm	1 x 10 ⁻² mm	1.0 x 10 ⁻⁴
WA Working Aperture	110 mm	1 x 10 ⁻² mm	9.1 x 10 ⁻⁵
WT Window Transmission	0.810	0.001	1.2 x 10 ⁻³
FT NDF Transmission	0.274	0.001	3.6 x 10 ⁻³
T CP Transmission	0.975	0.005	5.1 x 10 ⁻³
CR Raw Readings	900	4.5	5.0 x 10 ⁻³

Combining the above values into equation 5 leads to a predicted relative uncertainty of: $U_{I_{cal}} = 0.008$

Relative Uncertainty of I_r: Under ambient operating conditions, the irradiance measured by the transmissometer receiver will fluctuate due to:

- atmospheric optical turbulence causing scintillation;
- atmospheric optical aberrations causing beam wander;
- varying meteorological conditions along path: rain, snow, fog; and
- insect swarms causing beam interference.

The precision of each average irradiance measurement is estimated by the standard deviation of the one-minute irradiance measurements in the averaging period; thus, a direct estimation of any atmospheric optical interferences is made. The relative uncertainty in I_r has been investigated in the National Park Service monitoring network. Typical values are:

$$\begin{aligned} \text{for non-weather affected data: } & U_{I_r} = 0.006 \\ \text{for weather affected data: } & U_{I_r} = 0.15 \end{aligned}$$

Relative Uncertainty of Fl_{amp}: The lamp brightness of the transmitter, while controlled by an optical feedback circuit, actually has a slight brightening function related to the amount of hours that the lamp has been in service. Detailed investigations of this effect have found that the typical lamp brightens at a rate of 2.0% per 500 hours of lamp life. The precision of these measurements has been about 0.2%. The relative uncertainty in Fl_{amp} as the lamp ages is: $U_{lamp} = 0.002$

Relative Uncertainty in Path Transmittance: Combing the above analyses, the relative uncertainty in path transmittance can be calculated. Typical values are:

	U_T
No Optical Interference	0.010
Optical Interference	0.20

Precision of Extinction Estimates From Transmittance Measurements: The average extinction (b_{ext}) of the optical path (r) is calculated from the path transmittance (T) by:

$$b_{ext} = - \ln(T)/r \quad (6)$$

The path length r is measured to an extremely high precision; thus, the precision in b_{ext} can be approximated:

$$\sigma_{b_{ext}} = \pm U_T/r \quad (7)$$

The relative uncertainty in transmittance leads to an additive uncertainty in extinction and the magnitude is dependent on the path length. The following lists the predicted uncertainties of b_{ext} estimates for the Shenandoah operational paths when optical interferences are and are not present:

Path	Uncertainty	
	WX Interfer.	No WX Interfer.
1.41km	0.14 km ⁻¹	0.007 km ⁻¹
0.67km	0.30 km ⁻¹	0.015 km ⁻¹

Bias In Extinction Calculations: The calibration equation assumes clean glass surfaces of constant transmittance. Any change in the window transmittance results in a bias added to the calculated extinction. If the window transmittance decreases the calculated extinction will increase, if it increases the calculated extinction will decrease. As with the precision, the bias is a function of the relative change in window transmittance and path distance:

$$\text{Bias} = (\text{relative change in window transmittance})/r \quad (8)$$

The possibility exists for errors to arise from changes in the transmittance of the windows due to:

- pitting of the windows by wind blown dirt;
- staining of the windows by pollution;
- dirt collecting on the window surface due to dust, rain, snow;
- fogging of the windows at high humidities;
- improper servicing resulting in smudging of the windows; or
- removal of the windows due to breakage.

National Park Service (NPS) transmissometer data collected during 1991 was used to investigate the bias associated with varying window transmittance. Field operators are instructed to visit both the receiver and transmitter weekly. One of their duties is to observe the windows carefully and clean them regularly. These actions are noted on field log sheets. The NPS data base was scanned to locate the indicated times when the windows of the transmissometer systems were cleaned. The previous three hours and the following three hours of data were extracted for each cleaning. Servicing periods when the measured irradiance was constant before the windows were cleaned and also remained constant (independent of the previous three hours) after cleaning were identified. 335 servicings were selected that met these requirements. The average change in window transmittance was calculated from the difference between the mean irradiance values before and after servicing from this data set. The mean change was found to be 0.1%. This is misleading due to the fact that the servicing of the windows can have three possible effects:

- No change in window transmittance - the windows were perfectly clean before and after servicing
- The window transmittance increased - the windows were dirty and servicing cleaned them
- The window transmittance decreased - the windows were clean and servicing made them dirty

The first condition leads to no change in window transmittance thus no bias. The second condition would indicate that b_{ext} values measured before the servicing were biased too high. The third condition would result in b_{ext} values measured after window cleaning biased to high. Thus, in practice, unless the window is removed or a window with a higher transmittance is substituted, the bias due to a change in window transmittance is in one direction: increasing the calculated extinction either before or after the servicing. If conditions 2 and 3 have about the same magnitude and occur at about the same frequency, a simple comparison of mean radiance differences before and after servicing will come out as a zero percent change. Therefore, a better indication of this bias is a calculation using the absolute value of the difference in mean radiances measured before and after servicing. When this is done, the mean change in window transmittance for the NPS network was 1.5%.

Integrating Nephelometers

The uncertainty in scattering measurements is due to electronic drift which manifests itself as a change in the slope of the calibration line defined by the clean air and Freon span values. Unlike transmissometers where an uncertainty in the transmittance leads to an additive error in extinction; the uncertainty of the slope of a nephelometer calibration line, leads to a percentage error that is a multiplier on all scattering measurements. The variance in the slope of the calibration line for ambient Belfort nephelometers has been investigated in a number of monitoring programs. Table 1 lists the uncertainty statistics that have been observed. The uncertainty of the Belfort systems is highly variable and extremely temperature dependent¹³, ranging from 10% to over 100%. The Optec NGN-1 used during the Shenandoah summer intensive had an uncertainty of 11%. The modified version (NGN-2) used during the 1992 MOHAVE study had an estimated precision of 5%. The improved precision is traceable to modifications in the flow characteristics of the sampling chamber and span gas system in the NGN-2. Additional laboratory tests have indicated that the uncertainties of 5% or less will be regularly obtained with the NGN-2 system during routine operation.

The linearity of the calibration line for the Optec NGN-1 nephelometer has been examined by using a number of span gases¹⁸. Table 2 lists the gases used and figure 4 is a plot of the theoretical scattering values of these gases as a multiple of Rayleigh (clean air) versus the measured (fifteen minute integration) values adjusted for the slope (electronic gain) and intercept (wall scattering) of the calibration line. The output of the nephelometer is highly linear and able to respond to Helium which has a scattering coefficient only 1.3% of Rayleigh. The linearity of this relationship has been examined in the laboratory under varying temperatures and found to be extremely stable.

TABLE 2

Theoretical Multiple of Rayleigh Scattering

Gas		Rayleigh Multiple
Freon 12	(F12)	15.31
Freon 22	(F22)	7.69
Carbon Dioxide	(CO2)	2.61
Rayleigh	(Air)	1.00
Argon	(Ar)	0.879
Helium	(He)	0.013

Relative Humidity Sensors

The precision of relative humidity sensors have not been tested in previous visual air quality studies. Published values collected from manufacturers have been accepted. The summer intensive examined the response of two capacitive type instruments versus an aspirated wet/dry bulb system. Figures 5 and 6 indicate that the Rotronic MP-100MF sensor exhibits a far better response over the complete range of humidities experienced than the Campbell 207. Table 3 lists the performance of the two thin-film capacitive sensors versus the wet/dry bulb system. The Campbell sensor has approximately twice (5.3%) the uncertainty of the Rotronics sensor (2.7%).

TABLE 3

Performance of Rotronics MP-100MF and Campbell 207 Relative Humidity Sensors Compared To Aspirated Wet/Dry Bulb (WD) Reference Sensor

Sensor	Number Points	Difference		Ratio	
		WD(rh)-Sensor(rh) Mean	Sigma	WD(rh)/Sensor(rh) Mean	Sigma
Rotronics	1940	0.65%	2.7%	0.990	0.035
Campbell	1938	-0.55%	5.3%	1.001	0.066

TRANSMISSOMETER DATA CODING

Identification of Meteorological and Optical Interferences That Affect the Calculation of bext from Transmission Measurements

The average extinction coefficient of the sight path is calculated from the measured path transmittance and is attributed to the average concentration of atmospheric gases and ambient aerosols along the sight path. The intensity of the light, however, can be modified not only by intervening gases and aerosols, but by the presence of condensed water vapor in the form of fog, clouds, and precipitation along the sight path or condensation, frost, or fog forming on the shelter windows. In addition to these meteorological effects, two other possible major sources of optical interference may exist:

- reduction in light intensity by insects, birds, animals, or vegetation along the sight path, or present on the optical surfaces of the instrumentation or shelter windows; or
- fluctuations in light intensity both positive and negative due to optical turbulence, beam wander, atmospheric lensing, and miraging caused by variations in the atmospheric optical index of refraction along the sight path.

A major effort was undertaken to develop an algorithm to identify transmissometer extinction data that may be affected by the interferences described above. This algorithm contains five major tests:

- 1) Relative Humidity;
- 2) Maximum Extinction;
- 3) Uncertainty Threshold;
- 4) Rate of Change of Extinction; and
- 5) Isolated Data Points

Due to the immense amount of extinction data collected by transmissometers as compared to aerosol monitors, the algorithm has been designed to be a conservative filter on the extinction data. That is, if an hourly extinction measurement indicates the slightest possibility of meteorological or optical interference by failing any one of the above tests, it is flagged with identifier codes in the Level-1 data file. The following describes each of the five tests:

Relative Humidity: When the relative humidity measured at the transmissometer receiver is greater than 90%, the data is flagged as having a possible interference. The 90% level has been chosen due to the following considerations:

- The RH is only measured at the receiver location and not at any other position along the sight path;
- A 1.5° C change in dew point temperature results in a 10% change in RH;
- The atmosphere is continuously undergoing both systematic and random variations in its spatial and temporal properties; and
- The typical precision of past RH measurements is only $\pm 5\%$.

The above considerations all indicate that inferring a precise knowledge of the meteorological conditions along a optical path at high relative humidity from a single point measurement of RH is very difficult. When the relative humidity is above 90% at one end of the path, small random temperature or absolute humidity fluctuations along the path can lead to condensation of water vapor causing meteorological interferences. Thus, in accordance with the conservative philosophy expressed above, the 90% relative humidity limit was selected.

Maximum Threshold: For every transmissometer path, a maximum bext can be calculated that corresponds to a 2% transmission for the path. Sight paths are selected, such that based on historical visibility data, this bext occurs less than 0.5% of the time. When the measured bext is greater than this value, it is assumed that meteorological or optical interferences, not ambient aerosols, are causing the high extinction, and is so flagged.

Uncertainty Threshold: A mean hourly extinction and standard deviation is calculated from the number of one-minute irradiance values collected during the sampling period. The uncertainty is then calculated as described earlier. Typically, the ambient aerosol concentration varies quite slowly with time constants on the order of a few hours rather than minutes. This leads to the expectation of relatively constant extinction during the period of receiver measurements and a low standard deviation of measured transmitter irradiance. If only a few of the irradiance values varies more than 20% from the mean, the uncertainty in bext will increase dramatically. The presence of any of the above meteorological or optical interferences along the sight path will lead to large standard deviations in lamp irradiance, thus large uncertainties in bext. With the

conservative assumption of constant bext during the measurement period, any increase in the uncertainty of bext above some threshold flags the measurement as affected by one of these interferences.

Rate Of Change Of Extinction (Delta Threshold): This test consists of comparing the hourly average extinction to the preceding and following hours, and calculating a rate of change in each direction. If the absolute value of this rate of change is greater than some assigned delta threshold, the hourly bext value is flagged as being affected by interferences. These delta thresholds have been evaluated for each sight path by analyzing extinction data collected to determine the appropriate delta thresholds. The delta threshold is typically not as low as the one set for the uncertainty threshold, due to the possibility of larger hourly variations in bext compared to variations during the shorter sampling period during the hour.

Isolated Data Points: This test is performed after the above four thresholds are applied to the hourly extinction data. It is used to identify data points that have passed the above thresholds, but are located between hourly bext data that have failed the above thresholds. The conservative assumption is, if data before and after the isolated hour indicates interferences, the hour in question probably is also affected by interferences. This data is also flagged as "weather-affected."

Level-1 Weather Codes: Level-1 hourly data files contain validity flags, called for simplicity, weather identification codes. Each hourly bext value is tested for the above five thresholds. If the value fails any one of the thresholds it is identified as weather-affected. In addition, separate codes identify the specific threshold or combination of thresholds that the bext value has failed. Figure 7 is plot of one month of transmissometer extinction data collected by the long-path instrument. The top section contains all the extinction data, the bottom line only shows the data that is considered to be unaffected by meteorological or optical interferences. Extinction data that has been edited for these effects has proved to be highly correlated with reconstructed extinctions from aerosol data at Shenandoah National Park.

MEASUREMENT INTERCOMPARISONS

Transmissometers - bext

Figure 8 is a scatter plot (with the one-to-one line indicated) of hourly extinction data collected by the two short-path transmissometers during the first six weeks of the summer study. After this period one of the systems was damaged by a lightning strike and the following data was highly suspect. Figure 9 is a similar plot of data collected by two transmissometers during a similar intercomparison study at Tonto National Monument in Arizona. These systems were operated side-by-side over the a very long (7.2 km) path. Both figures indicate the extremely high precision of transmissometers to replicate extinction measurements when operating over identical paths. Figure 10 is a scatter plot of short-path bext vs. long-path bext during the summer study. The correlation is again outstanding. Analysis of the extinction data from the short- and long-paths indicate that the predicted uncertainties listed previously for weather and non-weather affected data agree very well with the actual calculated uncertainties:

Path Length	Hours of Data			ALL Data		Non-Weather Data	
	All	Non-WX	%	Mean bext	Uncertain.	Mean bext	Uncertain.
1.41 km	2174	1472	68	0.87	0.13	0.214	0.011
0.67 km	2216	1307	59	0.94	0.27	0.207	0.015

Nephelometers - bscat

Figure 11 is a comparison of hourly averaged bscat measured by the Optec and ambient Belfort nephelometers. The correlation between the two instruments is very good. In fact, due to this correlation, the heating of the Optec nephelometer was investigated very thoroughly after the field study. It was determined that unacceptable heating of the ambient aerosol was occurring and the modifications discussed previously were incorporated into the second generation Optec NGN-2 nephelometer.

Figure 12 is a comparison of the Optec versus heated inlet Belfort nephelometer. It is quite obvious that even a small heating of 5.5° C leads to a dramatically lower bscat measurement. The increased scatter between ambient and heated bscat measurements is due to the variability of hygroscopic characteristics of the ambient aerosols. Thus, the same heating will have a different effect on bscat depending on the amount of liquid water condensed on the particles. The mean bscat ratios are:

$$\begin{aligned} \text{Belfort ambient/Optec} &= 0.997 \\ \text{Belfort heated/Optec} &= 0.63 \end{aligned}$$

Recalling that both ambient nephelometers heated the aerosol by about 1.5° C and that the heated cycle warmed the aerosol by 5.5° C; there is an amazing mean reduction in measured bscat of about 37% due to the 4° C warming by the heated inlet over the ambient cycle. This only serves to strongly verify the fact that any heating of the ambient aerosol during a scattering measurement at high relative humidities will seriously underestimate the true ambient scattering coefficient!

Transmissometer Bext vs Nephelometer Bscat

Figure 13 shows the correlation between hourly mean bext as measured by the long-path transmissometer and hourly mean bscat by the Optec NGN-1 nephelometer at relative humidities below 70%. The correlation is very good. The slope of the best fit least squares line to the data is 1.19 with an intercept of -0.002. This indicates that the transmissometer measured bext is about 19% higher than the bscat measured by the NGN nephelometer. The higher measured extinction compared to scattering is caused by a combination of aerosol absorption and an underestimate of bscat by the nephelometer due to the 1.5° C heating of the ambient aerosol by the system.

Figure 14 is a similar plot of bext versus bscat, but for relative humidities between 70% and 90%. The correlation drops dramatically. The increased scatter is due to a number of considerations:

- At higher relative humidities heating the aerosol causes a greater under-estimation of bscat;
- The variability in the concentration of hygroscopic aerosols causes non-uniform reductions in bscat with the same heating by the nephelometer;
- At higher relative humidities and higher pollution levels the effect of the separation between the transmissometer sight path and the nephelometer location becomes more pronounced. The atmosphere is more "blobby". The non-uniformity results in fairly large differences ambient extinction levels at locations only a few kilometers apart.
- The same effects are seen along the transmissometer sight path resulting in non-uniform extinction along the path. Pockets of high extinction air either due to aerosols or high humidity "blobs" pass across the sight path decreasing the transmittance of the path.

Employing a longer averaging period reduces the effect of non-uniformity of the atmosphere. However, the effect of heating on varying hygroscopic aerosol concentrations remains and is not removed by increasing the averaging period.

CONCLUSIONS

The following conclusions concerning ambient optical measurements are supported by this work:

Transmissometers:

1. Extinction data from transmissometer measurements are truly ambient in nature, highly precise, and accurate;
2. The uncertainties associated with transmissometer data are understood and quantifiable from pre- and post-calibrations, collected irradiance data, and operating parameters;
3. Bias in transmissometer measurements can be estimated from operating protocols and an examination of extinction data;
4. Meteorological and optical interferences in extinction data from transmissometer measurements can be identified with the use of algorithms that examine the precision and consistency of the collected data.

Belfort Nephelometers

1. Belfort nephelometers can be operated in a near ambient mode; however, the lowest chamber-ambient heating possible is about 2° C;
2. Operating Belfort nephelometers near ambient conditions is expensive and leads to increased uncertainty in the scattering measurements;
3. Optical chamber temperatures of any nephelometer must be monitored to correct the scattering data for heating of the ambient aerosol; previously collected nephelometer data that does not have associated optical chamber temperatures must be considered highly suspect;
4. Care must be taken when operating the nephelometer in a switching heated/ambient mode to be sure that the system has come to temperature equilibrium before using the bscat data in any averaging period.

Optec NGN Nephelometers

1. The nephelometer output is highly linear and stable under varying temperatures, thus exhibiting high precision;
2. Scattering data collected by the first prototype correlated quite well with the Belfort ambient system at relative humidities below 90% and transmissometer measurements of bext at relative humidities below 70%;
3. The first prototype exhibited similar aerosol heating as the Belfort ambient system; improvements in the system have lowered the measured ambient optical chamber temperature differences to around than 0.5° C;
4. The system proved to be rugged and reliable while operating continuously during the intensive monitoring period.

Relative Humidity Sensors

1. The new Rotronics relative humidity sensor exhibits a far better linear response with twice the precision over a wider range of ambient humidities than the Campbell 207;
2. Historic relative humidity data collected with older sensors similar to the Campbell 207 should be used with caution.

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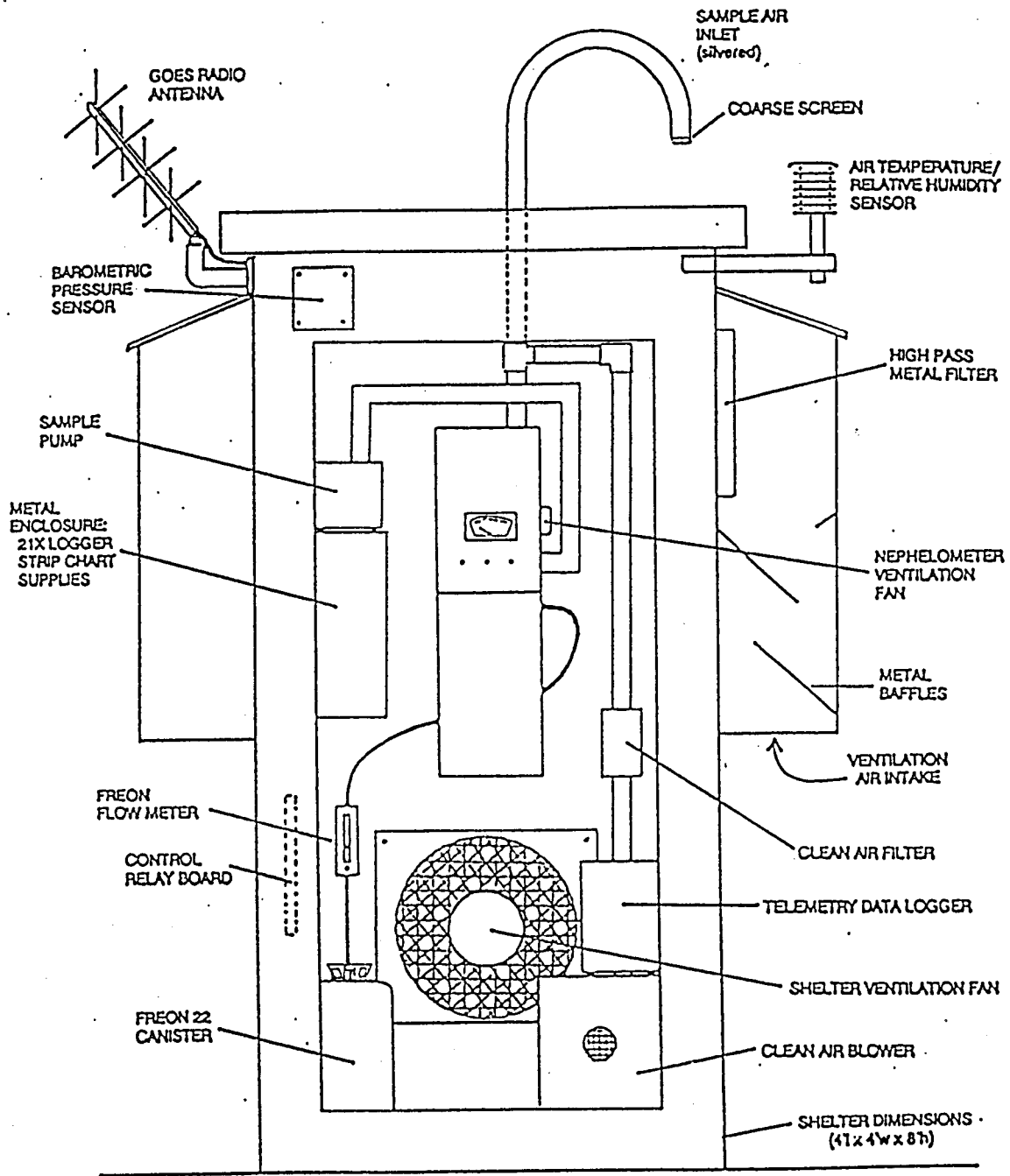


Figure 1. Belfort Ambient Nephelometer System Functional Diagram

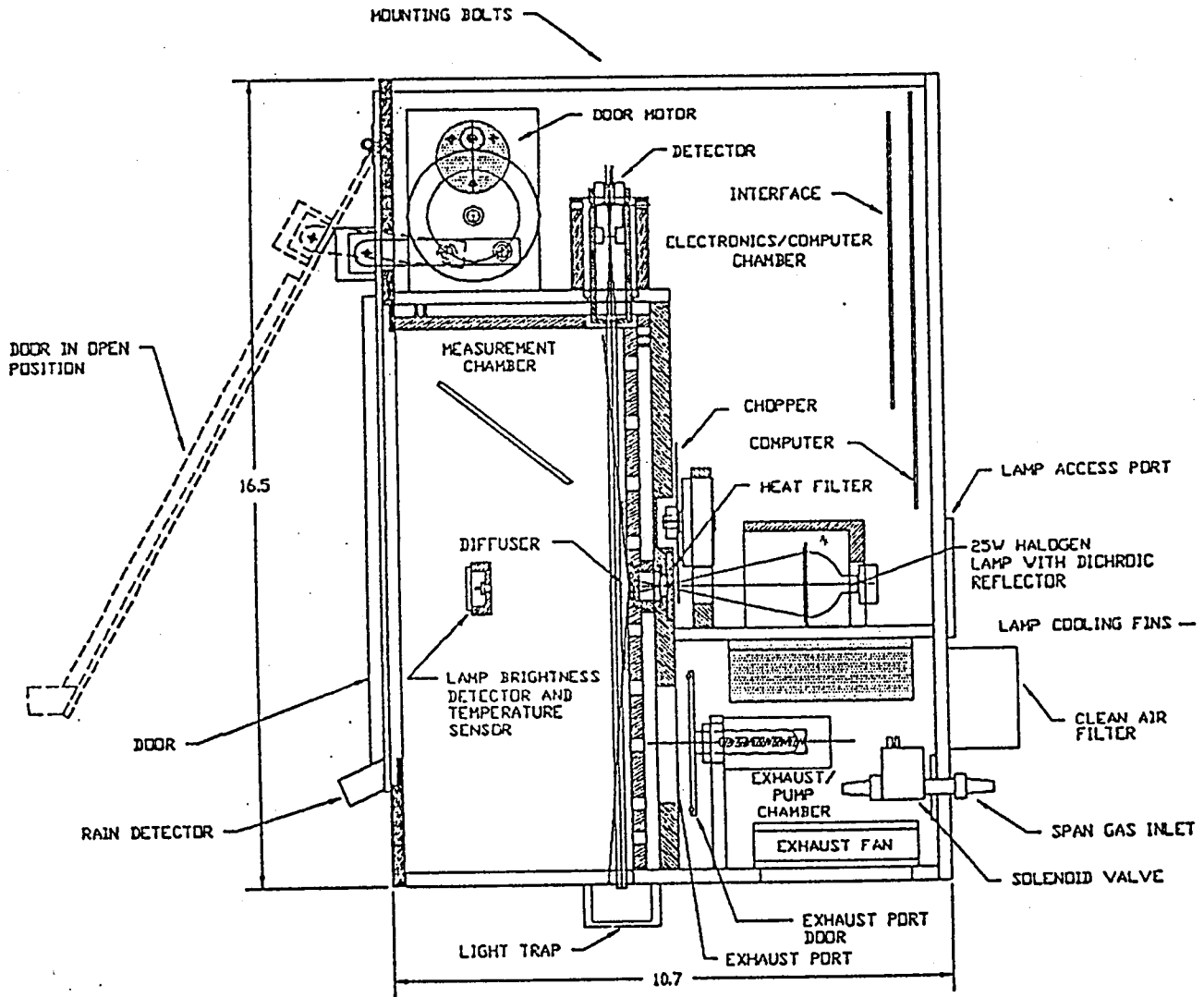


Figure 2. Optec, Inc. NGN-1 Nephelometer Functional Diagram

Shenandoah National Park
Belfort Neph b_{scat} and Temperatures

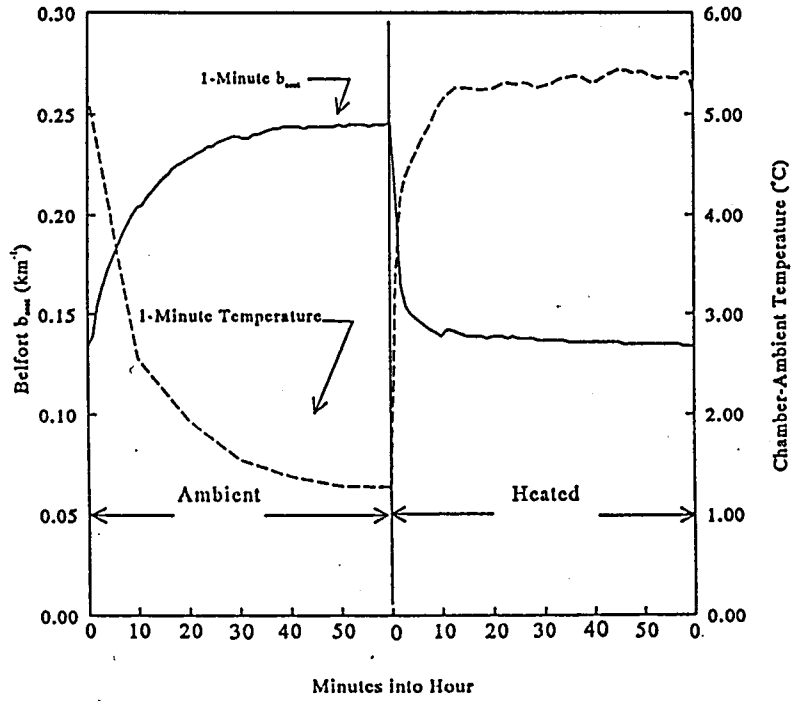


Figure 3. Mean b_{scat} and Temperature Response Belfort Switching Nephelometer

Measured vs. Theoretical
Span Gas Scattering

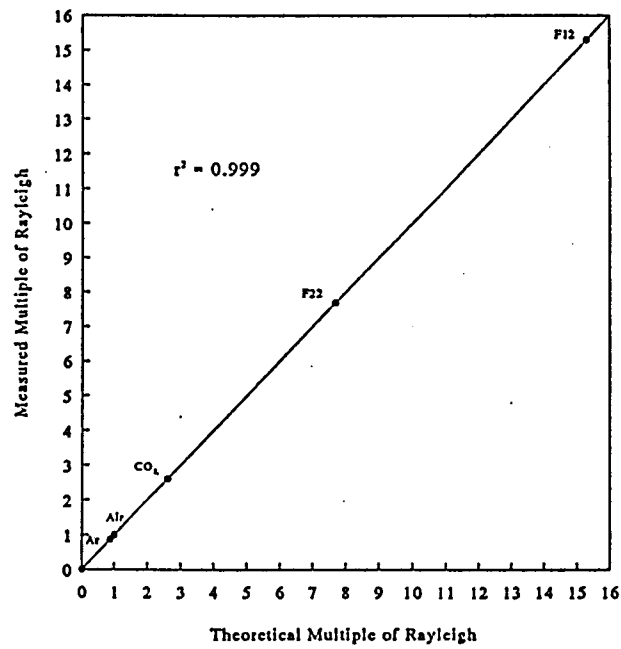


Figure 4. Optec, Inc. NGN-1 Span gas Calibration

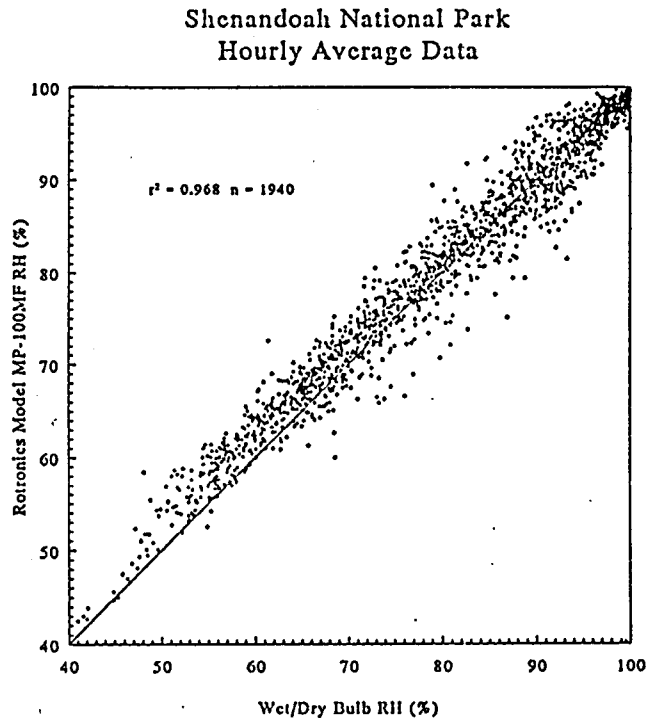


Figure 5. Rotronics MP-100MF vs. Wet/Dry Bulb Relative Humidity

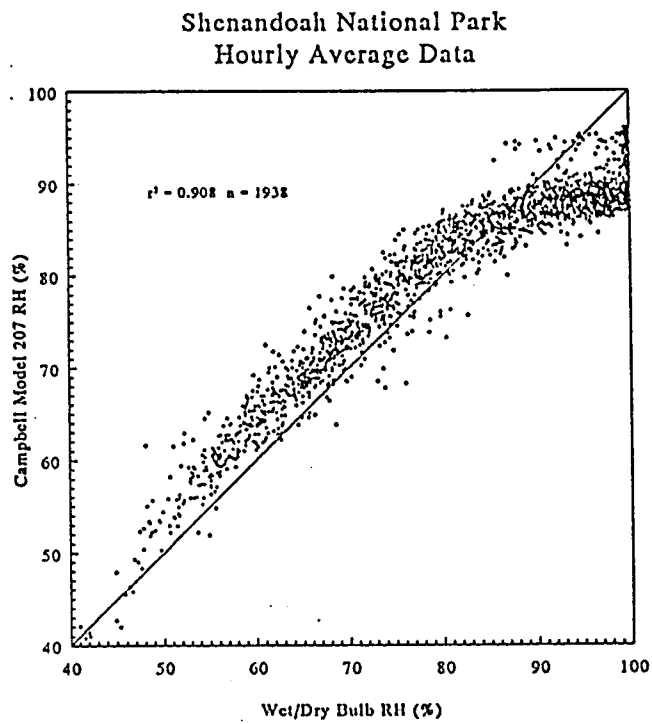


Figure 6. Campbell 207 vs. Wet/Dry Bulb Relative Humidity

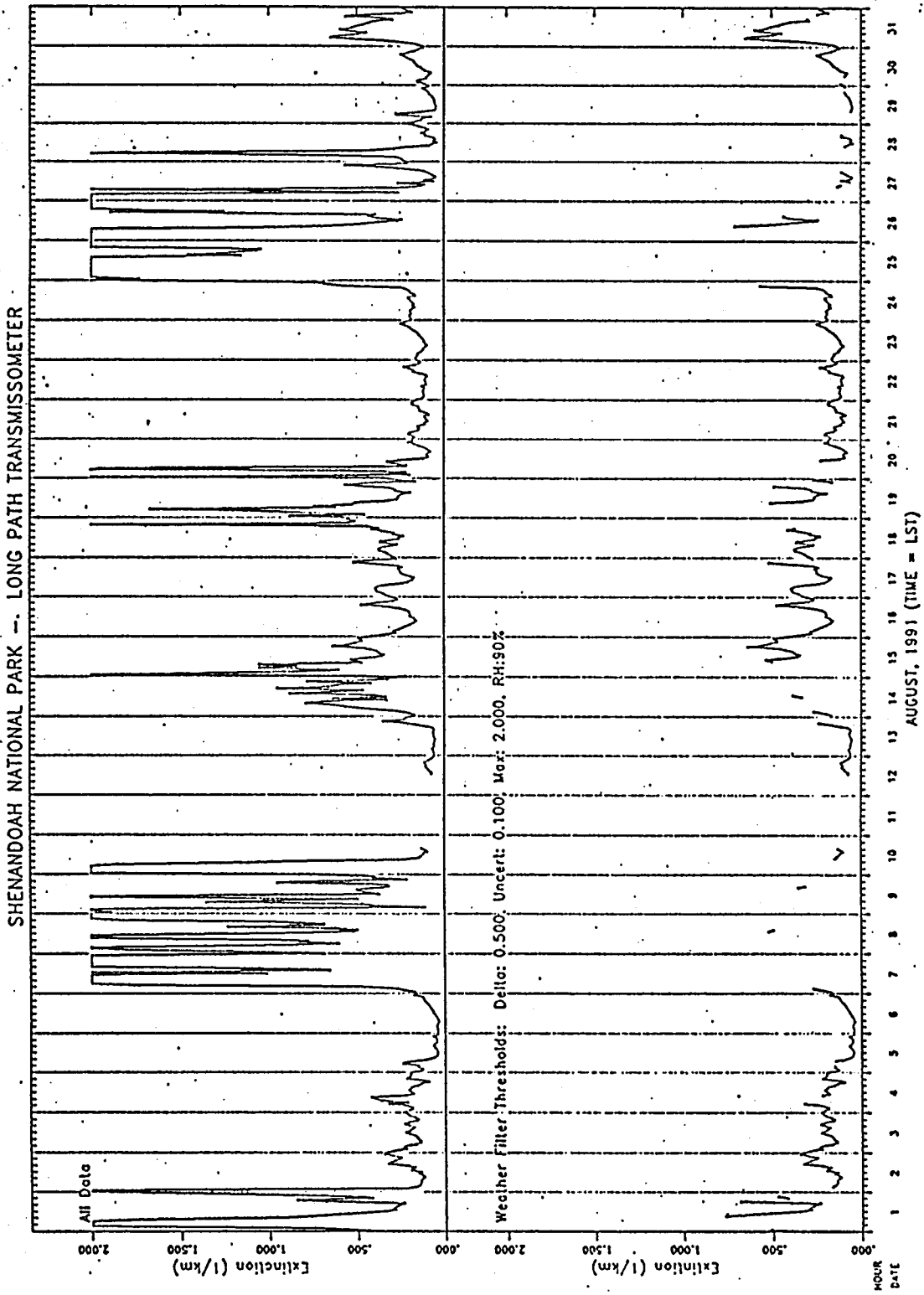
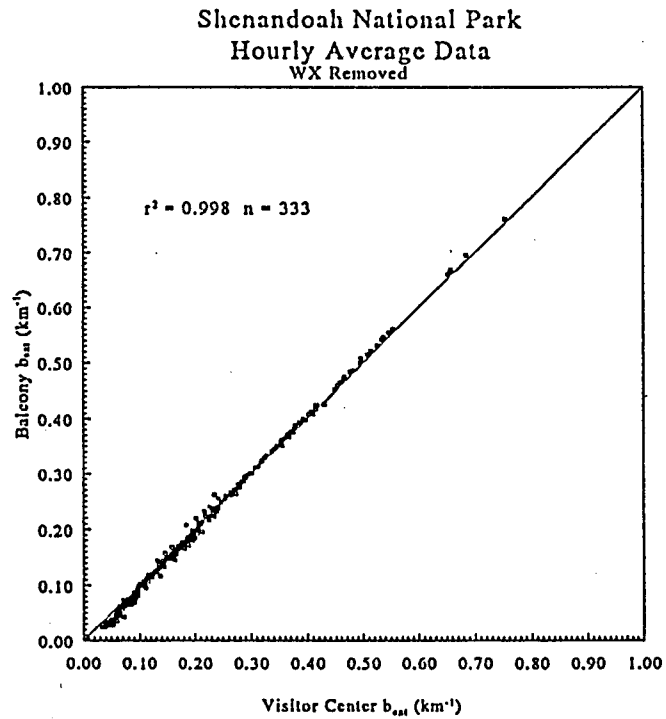
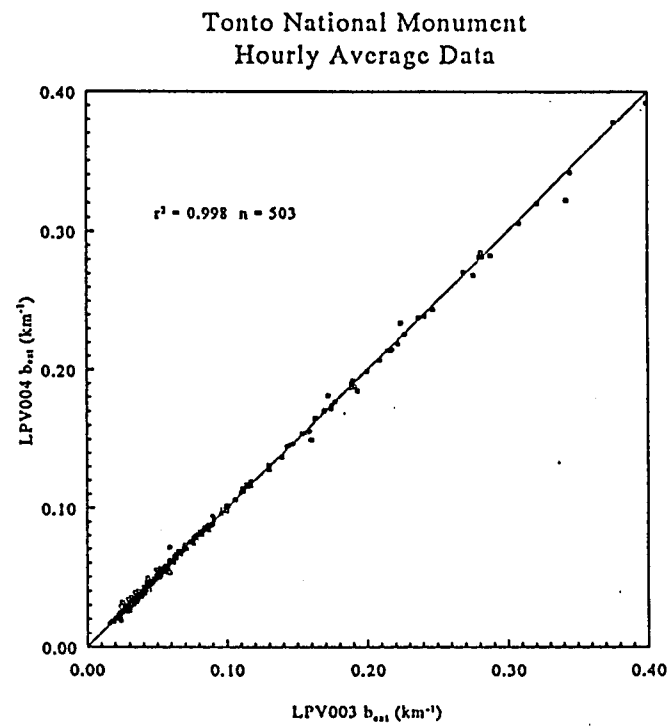


Figure 7. Application of Weather Algorithm

Figure 8. Short Path (0.67 km) b_{ext} ScatterplotFigure 9. Long Path (7.2 km) b_{ext} Scatterplot

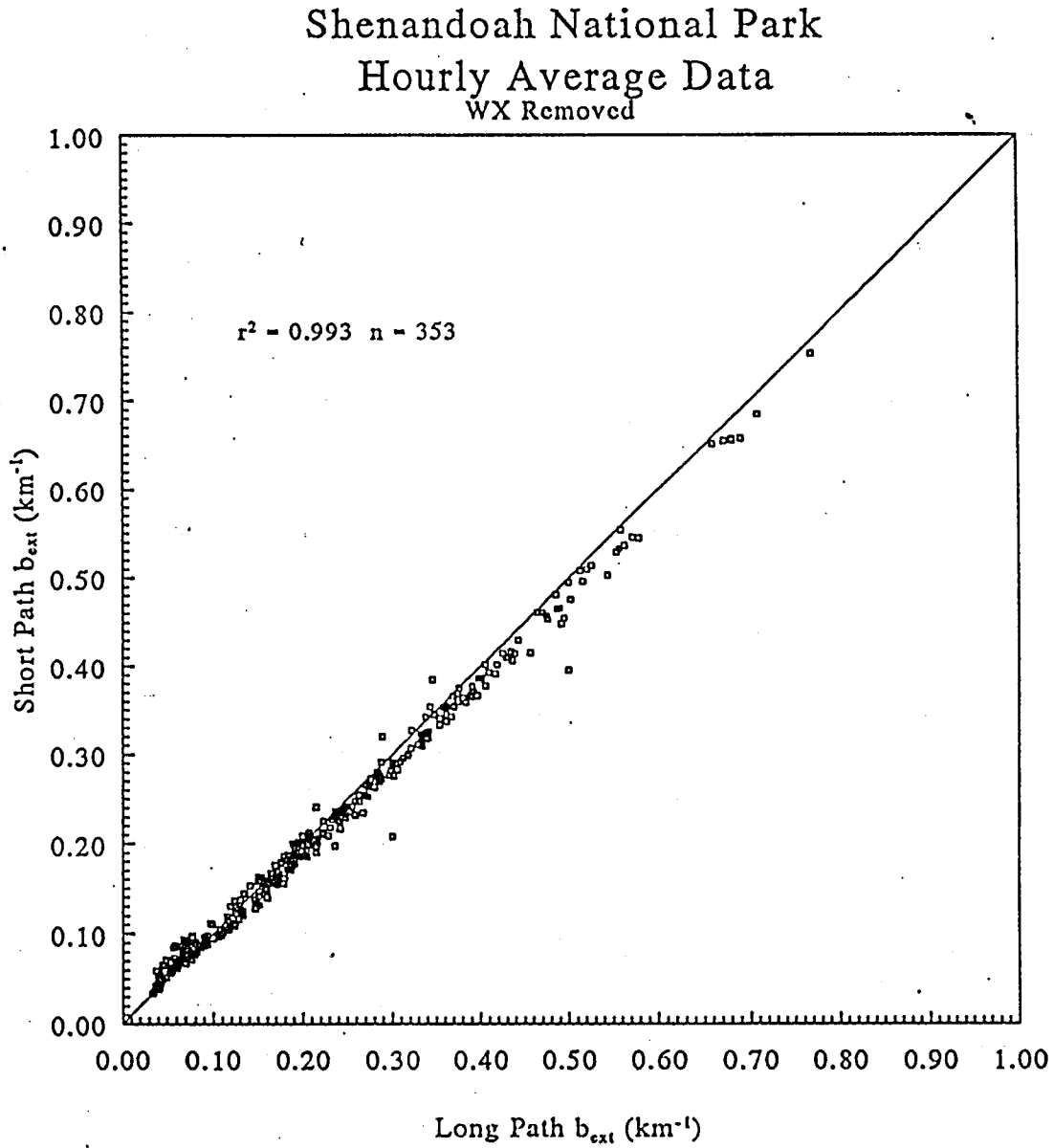


Figure 10. Short Path (0.67 km) vs. Long Path (1.41 km) b_{ext} Scatterplot

Shenandoah National Park
Hourly Average Data

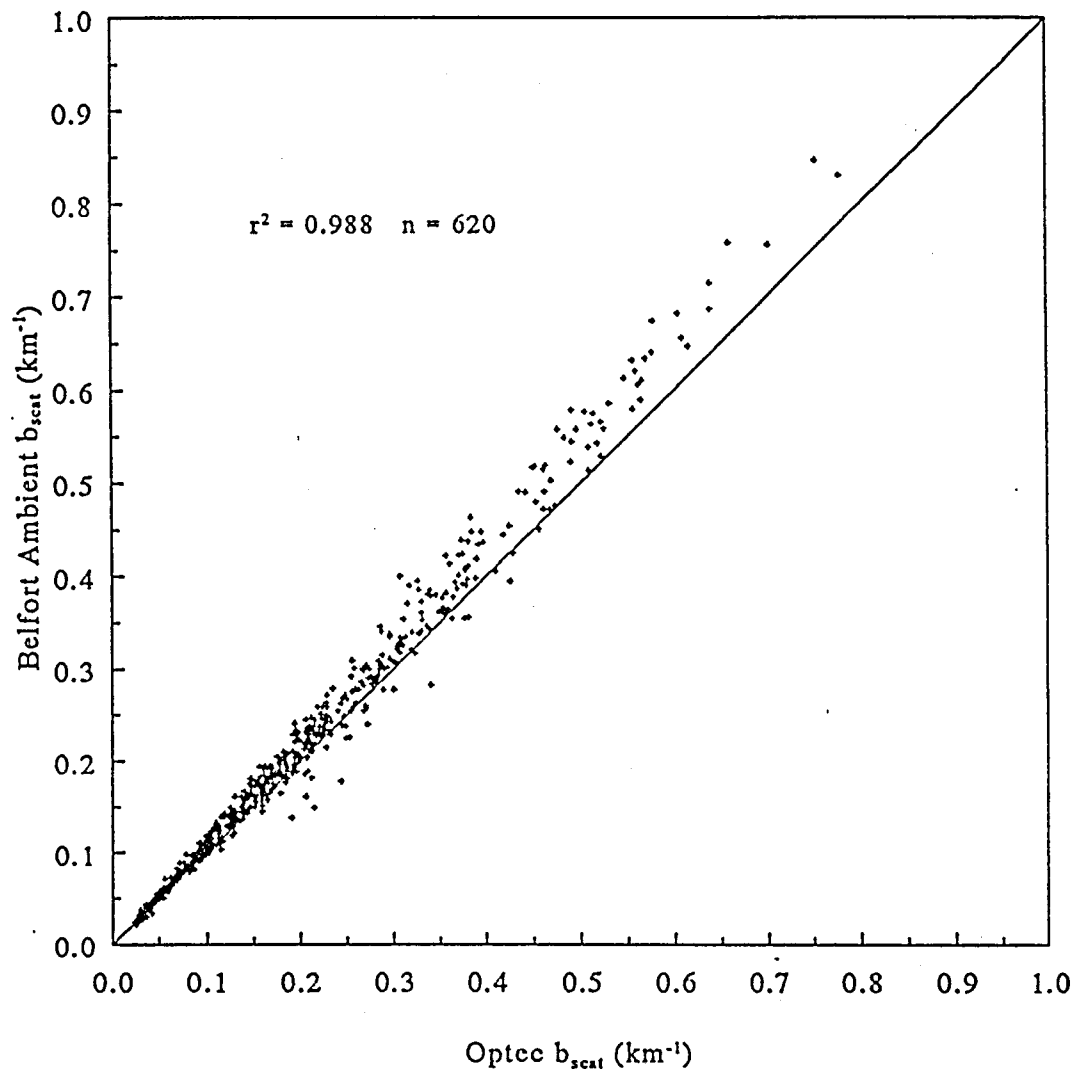


Figure 11. b_{scat} : Belfort Ambient vs. Optec $rh < 90\%$

Shenandoah National Park
Hourly Average Data

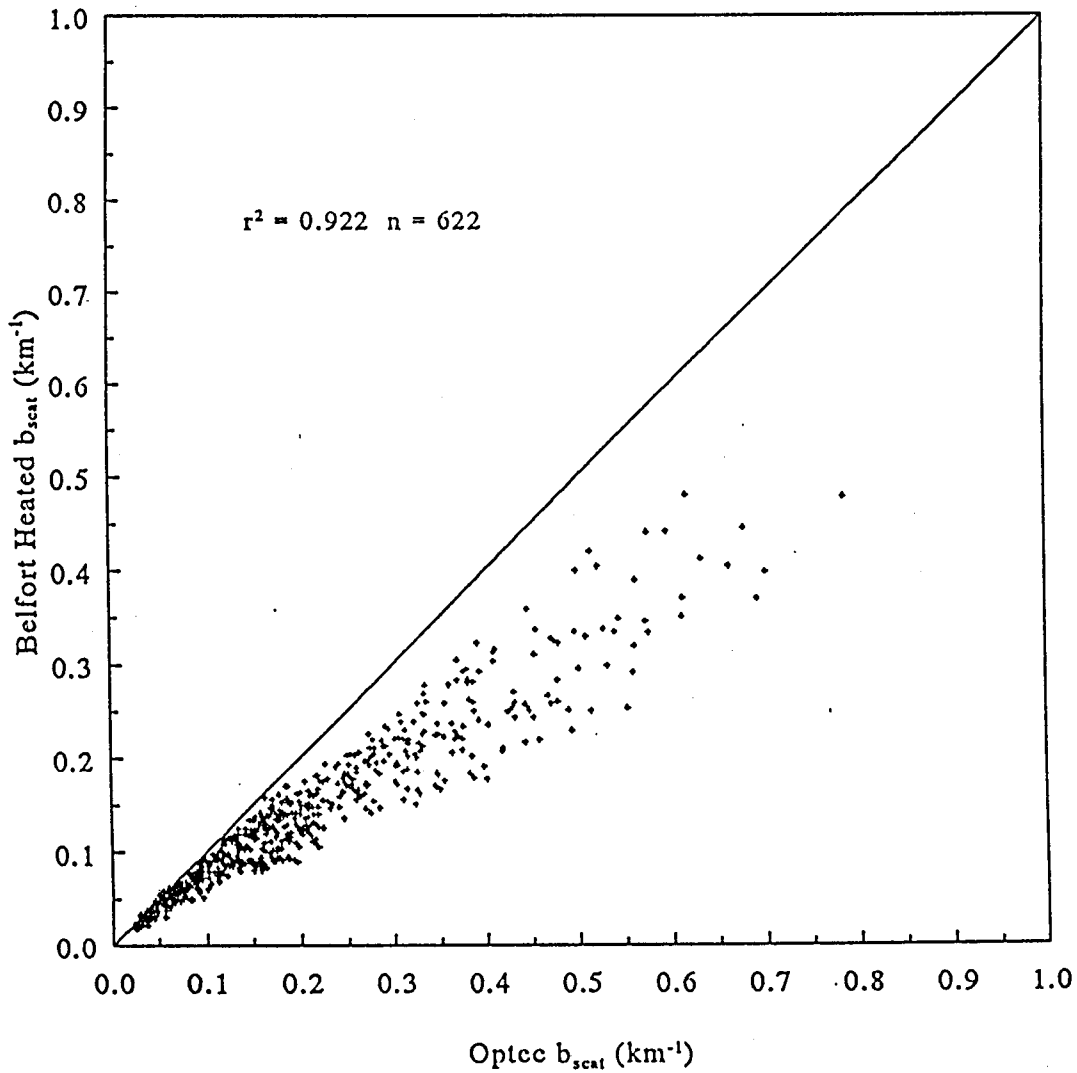


Figure 12. b_{scat} : Belfort Heated vs. Optec $rh < 90\%$

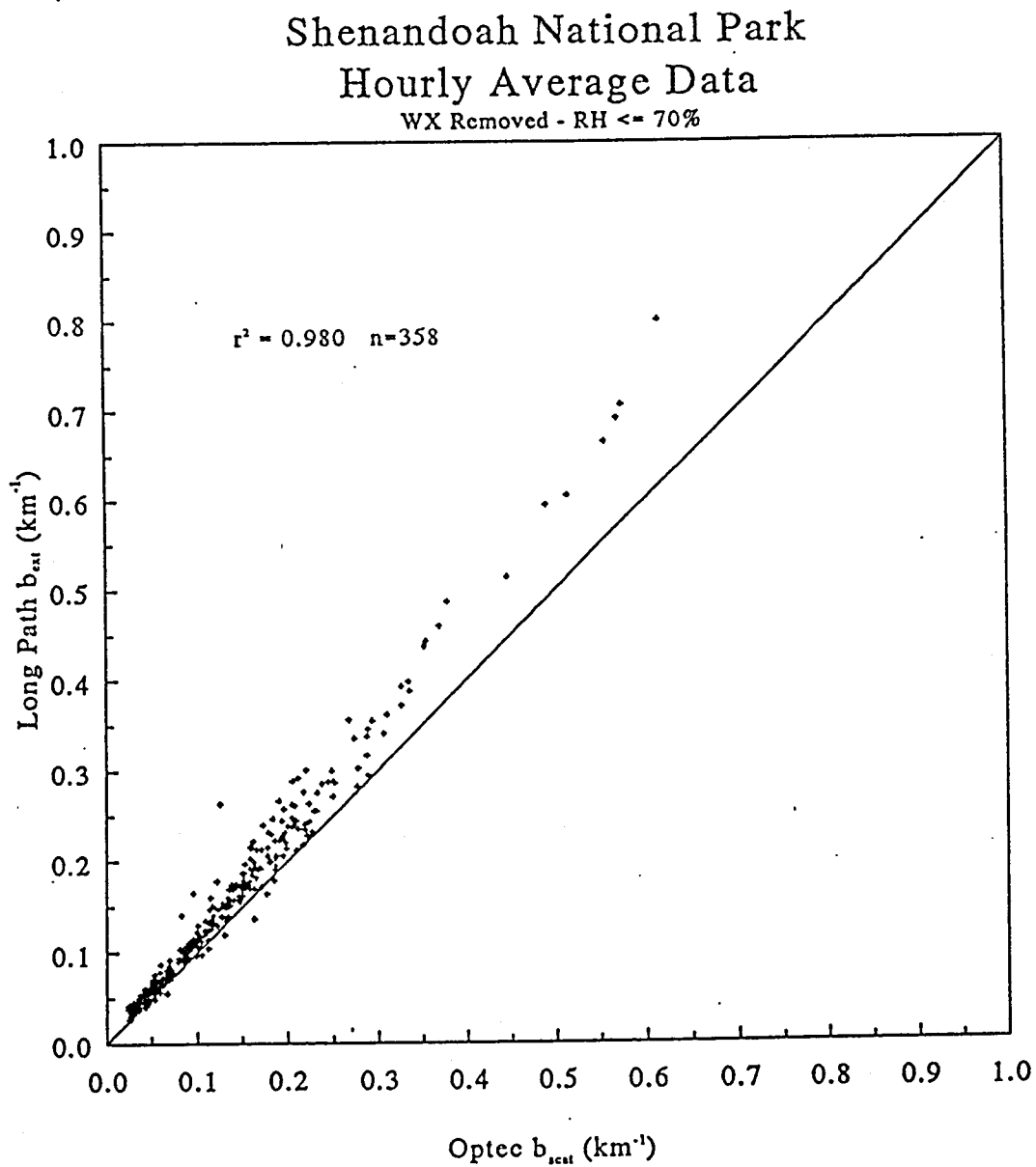


Figure 13. b_{ext} versus b_{scat} $rh \leq 70\%$

Shenandoah National Park
Hourly Average Data

WX Removed - 70% < RH < 90%

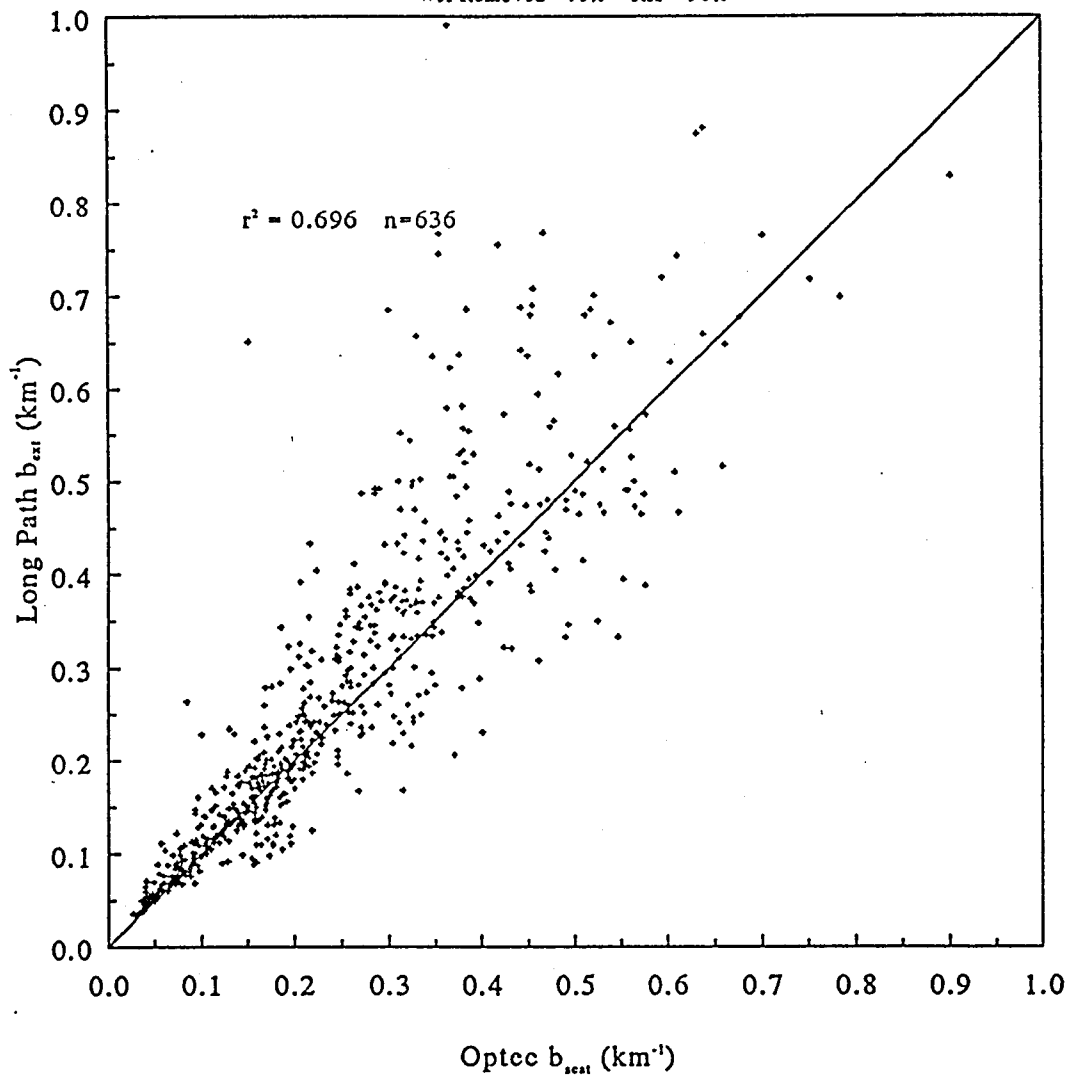


Figure 14. b_{ext} versus b_{scat} 70% < rh < 90%