Assessment of the UV camera sulfur dioxide retrieval for point source plumes

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ABSTRACT

Digital cameras, sensitive to specific regions of the ultra-violet (UV) spectrum, have been employed for quantifying sulfur dioxide (SO2) emissions in recent years. The instruments make use of the selective absorption of UV light by SO2 molecules to determine pathlength concentration. Many monitoring advantages are gained by using this technique, but the accuracy and limitations have not been thoroughly investigated. The effect of some user-controlled parameters, including image exposure duration, the diameter of the lens aperture, the frequency of calibration cell imaging, and the use of the single or paired bandpass filters, have not yet been addressed. In order to clarify methodological consequences and quantify accuracy, laboratory and field experiments were conducted. Images were collected of calibration cells under varying observational conditions, and our conclusions provide guidance for enhanced image collection. Results indicate that the calibration cell response is reliably linear below 1500 ppm m, but that the response is significantly affected by changing light conditions. Exposure durations that produced maximum image digital numbers above 32 500 counts can reduce noise in plume images. Sulfur dioxide retrieval results from a coal-fired power plant plume were compared to direct sampling measurements and the results indicate that the accuracy of the UV camera retrieval method is within the range of current spectrometric methods.

1. Introduction

SO2 emission measurements are an important component of monitoring volcanic processes, providing insight into the driving forces behind eruptions. Remote sensing techniques have been used for decades to investigate degassing and infer changes in the state of volcanic unrest. The standard ultra-violet (UV) ground-based remote sensing methods have used correlation (e.g., Stoiber et al., 1983) and differential optical absorption spectrometry (e.g., Galle et al., 2002; Horton et al., 2006). Imaging of SO2 has been accomplished at stack plumes (Exton, 1976), and in recent years, UV imaging systems have been applied to volcanic research (e.g., Mori and Burton, 2006; Bluth et al., 2007). These instruments have limitations in the spectrometric range, but are relatively inexpensive, address some of the limitations of earlier methods, and provide unprecedented insights into plume dynamics and emission rates at high temporal resolution (Yamamoto et al., 2008).

The UV camera components currently used by this research group are the Apogee Alta U6 camera with the Kodak KAF-1001E-2 1024×1024 pixel CCD, the Coastal Optics 105 mm UV SLR lens, and two Andover Optics bandpass filters. The filters are centered at 307 nm and 326 nm, and both filters have a band-width of 10 nm at half the maximum response (FWHM). The principle of the UV camera technique remains the same as for earlier UV remote sensing methods: the application of the Beer–Lambert law to determine the concentration of an absorbing species. UV images can be taken with just the 307 nm filter, which is centered in the SO2 absorbing region of the electromagnetic spectrum (Bluth et al., 2007) or with both the 307 nm filter and 326 nm filter (Mori and Burton, 2006). The 326 nm filter is centered in a relatively SO2 non-absorbing region of the spectrum and the contrast between the two-filter images has the effect of isolating the SO2 absorption feature while reducing the observed reduction in transmission due to other species (e.g., ash and aerosols). The UV images, which begin as arrays of illumination values (represented by digital numbers, DN), are converted to absorbance values using the Beer–Lambert law:

\[ A = -\log_{10}(I / I_0) \]

where \( A \) = absorbance, \( I \) = light intensity after passing through SO2, and \( I_0 \) = light intensity before passing through SO2 or background
light intensity. This method for converting relative light intensity values to absorbance using a single filter is the same as described in Bluth et al. (2007). The absorbance calculation used for the double filter method is a variation on that described in Mori and Burton (2006), the differences being that dark images were not used in the calculation (to facilitate a higher sampling rate) and the offset was not automatically removed in order to evaluate changes in this parameter. To translate absorbance to SO$_2$ pathlength concentrations, images of calibration cells (cells of known SO$_2$ pathlength concentration) are collected and the relationship between calculated absorbance and concentration is modeled (Fig. 1).

The benefits of the UV camera technique for imaging volcanic plumes are many: new insight into plume dynamics from two-dimensional measurements (Yamamoto et al., 2008); an internally-derived plume speed obtained from image sequences; the ability to retrieve small-scale speed obtained from image sequences; the ability to retrieve small-scale plumas; and comprehensive plume images simultaneously; and the high temporal resolution of the resulting dataset. However, the novelty of the technique can mean that procedural and instrumental limitations, procedural sources of error, and the accuracy of the method remain unknown.

When collecting measurement images with the UV camera, user-controlled parameters include image exposure length, the diameter of the lens aperture, the regularity of calibration cell imaging, the use of the single 307 nm filter or both the 307 nm and 326 nm filter, and if both filters are used, whether different or identical exposure times are used for each filter image. The initial field campaigns with UV camera instruments have led to inquiries into the amount of error introduced by variations in these parameters. We have conducted experiments into some of the user-controlled variables in the UV imaging technique, and provide some conclusions about optimal methodology. Additionally, we address the overall accuracy of the retrieval method and investigate some instrumental sources of error. Our study focuses specifically on the factors affecting the ability of the camera to correctly retrieve SO$_2$ concentrations; we do not address the methodology of converting concentrations to flux rates as described in Bluth et al. (2007).

2. Methods

In order to evaluate the UV camera for observing volcanic SO$_2$ emissions, the technique was assessed in three ways: instrument limitations, procedural sources of error, and field testing for accuracy. Instrument effects include sources of error or restrictions on the method due to the UV camera equipment. Sources of potential inaccuracy that stem from the user’s methodology in image collection and processing are addressed in Section 2.2. Finally, we tested the SO$_2$ retrieval accuracy in the field by comparing UV camera results to direct sampling data at a power plant. Table 1 describes the experimental design of each of these investigations.

2.1. Instrument limitations

The method for converting UV camera images from absorbance values to estimations of SO$_2$ pathlength concentrations requires imaging calibration cells (Fig. 1). The relationship between these calibration cell absorbance values is then applied to the plume image absorbance values. Three experiments were conducted to evaluate this procedure for accuracy and limitations. The first experiment was designed to appraise the calibration cell absorption response. Under laboratory conditions, UV camera images were collected of a magnesium fluoride pressure cell at 14 different concentrations of SO$_2$ ranging from 0 to 6048 ppm. Second, to address the effect of changing image background on the retrieved SO$_2$ concentration and determine precision in calculated absorbance, six images were taken at a constant concentration (814 ppm) under varying weather conditions. Finally, field measurements were taken of calibration cells at 90 ppm and 270 ppm with the UV camera under variable imaging conditions and at two different field sites. The calibration curves determined from these images were compared to evaluate reliability of the absorption relationship.

From the first observations made with the UV camera, it was clear that plume images suffered from a vignetting effect — a darkening towards the corners of the image. Vignetting is caused by different mechanisms, and the correction is dependent on the type of vignetting. Optical vignetting can be diminished by reducing the lens aperture, whereas natural vignetting is a function of the lens design and cannot be rectified by decreasing the aperture diameter (Ray, 2002; van Walree, 2007). To characterize the UV camera vignetting, images were taken of calibration cells at 7 different aperture settings, ranging from approximately 3.3 mm to 23.3 mm in diameter (Table 2). All images were taken at exposure times that resulted in brightness values that approached the maximum recordable values at the well depth and digital resolution for this
The light intensity values recorded by the UV camera are highly affected by the exposure time selected for the CCD. Field conditions generally determine the exposure time; under low light conditions a longer exposure time will allow more information to be recorded, but will blur the plume and decrease the SO₂ flux sampling rate. When conditions are too bright, the camera sensor can saturate resulting in information loss, so a shorter exposure time can be used. However, the camera shutter has a maximum speed and shorter exposure times will lead to shutter artifacts in the images. These factors lead the user to adjust the exposure time, but it is necessary to characterize the effects of such adjustments on the calculated SO₂ pathlength concentration. To test the consequence of exposure time on observed SO₂ absorption, sequences of UV images, including 5 calibration cells (0 to 1388 ppm m), were collected at a range of exposures with both the 307 nm and 326 nm filters. Exposure time is directly related to the light intensity (DN) recorded by the UV camera, and since exposure time is highly dependent on field conditions, the exposure times were selected to yield maximum intensity values ranging from approximately 4000 to 65000 counts for the 307 nm filter and similar values for the 326 nm filter images. The best-fit lines for the calibration cells at each exposure were then compared.

The UV camera can be used with a single 307 nm filter, or by comparing images taken with the 307 nm filter and the 326 nm filter to isolate the SO₂ absorption feature. A variable in the latter procedure is the use of “optimal” or “identical” exposure times for each filter and the 326 nm filter differencing technique. Optimal exposure times for each filter were determined by testing the number of counts recorded by the UV camera. Relative exposure times, “optimal” or “identical” exposure times for each filter, and similar values for the 326 nm filter images, are used to characterize the effects of such adjustments on the calculated SO₂ pathlength concentration. To test the consequence of exposure time on observed SO₂ absorption, sequences of UV images, including 5 calibration cells

### Table 1
Experimental design and parameters.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Location</th>
<th>Fixed variable</th>
<th>Fixed variable value</th>
<th>Free variable</th>
<th>Free variable range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Instrumental Calibration cell absorbance response</td>
<td>Resonance, Ltd., Ontario, CA</td>
<td>SO₂ source, background, light level, exposure time, aperture</td>
<td>MgF₂ pressure calibration cell, reflective board, UV lamp, 15 s (307 nm filter) and 3 s (326 nm filter), 23.3 mm</td>
<td>SO₂ concentration</td>
<td>14 values: 0 to 6048 ppm m</td>
</tr>
<tr>
<td>Uniformity of image background</td>
<td>Resonance, Ltd., Ontario, CA</td>
<td>SO₂ source, concentration, max image DN, aperture</td>
<td>MgF₂ pressure calibration cell, 814 ppm m, 30–56k counts, 23.3 mm</td>
<td>Weather conditions</td>
<td>Partly cloudy to snow flurries</td>
</tr>
<tr>
<td>Variability in concentration–absorbance relationship</td>
<td>Santiaguito and Pacaya Volcanoes, Guatemala</td>
<td>SO₂ concentration, image max DN</td>
<td>90 and 270 ppm m, &gt;30,000 counts</td>
<td>Light quality (image location and time of day)</td>
<td>2 field locations, 8:15–14:20</td>
</tr>
<tr>
<td>Image vignetting</td>
<td>Cascades Volcano Observatory, Vancouver, WA</td>
<td>Target, max image DN, image locations sampled</td>
<td>5 cal. cells (0 to 1388 ppm m), ~50000 counts, image corner, cal. cells, and transect</td>
<td>Lens aperture</td>
<td>3.3 to 23.3 mm in diameter</td>
</tr>
<tr>
<td>Procedural Exposure time significance</td>
<td>Cascades Volcano Observatory, Vancouver, USA</td>
<td>Target, aperture, filter method</td>
<td>5 cal. cells (0 to 1388 ppm m), 13.1 mm, single 307 nm filter</td>
<td>Exposure time (max image digital number, DN)</td>
<td>0.2–3.5 s (4000 to 65,000 counts)</td>
</tr>
<tr>
<td>Double filter method exposure time effects</td>
<td>Resonance, Ltd., Ontario, CA</td>
<td>Target, aperture, observational conditions</td>
<td>7 cells (0–1296 ppm m), 23.3 mm, laboratory environment</td>
<td>Relative exposure times</td>
<td>“Optimal” or “identical”</td>
</tr>
<tr>
<td>Field testing Power plant calibration</td>
<td>Wisconsin, USA</td>
<td>Plume concentration</td>
<td>435.0–450.1 ppm</td>
<td>Measurement method</td>
<td>Direct sampling (CEMS) and UV camera</td>
</tr>
</tbody>
</table>

### Table 2
F-stop numbers used in experimentation and corresponding aperture diameters for the Coastal Optics 105 mm UV SLR lens.

<table>
<thead>
<tr>
<th>F-stop designation</th>
<th>Aperture diameter (mm)</th>
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</thead>
<tbody>
<tr>
<td>F/4.5</td>
<td>23.33</td>
</tr>
<tr>
<td>F/5.6</td>
<td>18.75</td>
</tr>
<tr>
<td>F/8</td>
<td>13.13</td>
</tr>
<tr>
<td>F/11</td>
<td>9.55</td>
</tr>
<tr>
<td>F/16</td>
<td>6.56</td>
</tr>
<tr>
<td>F/22</td>
<td>4.77</td>
</tr>
<tr>
<td>F/32</td>
<td>3.28</td>
</tr>
</tbody>
</table>

**Fig. 2.** Location of brightness value measurements (indicated by dotted lines) for vignetting experiments. The values of the calibration cells were also evaluated for changes with variation in aperture diameter.
time is a single compromising exposure time intermediate to the two optimal exposure times, which allows for the longest 307 nm filter exposure without over-saturating the 326 nm filter (often equal to the optimal exposure time of the 326 nm filter). To determine the accuracy of each method, images of calibration cells were collected using both methods and under similar observational conditions. These images were processed to obtain best-fit lines; the slopes of these lines were evaluated as a measure of each method’s sensitivity to changes in SO$_2$ concentration.

2.3. Field testing

The accuracy of the UV camera technique was tested at a coal-burning power plant in Wisconsin, USA in August 2006. The field tests were completed on the same day as the plant RATA (relative accuracy test audit), ensuring that the continuous emission monitoring system (CEMS) data were correct for the stack emissions of SO$_2$. The CEMS measurements were taken from a sample probe centered in the stack exit duct. UV images of the SO$_2$ plume were collected using the 307 nm filter from three locations — 150, 1300, and 1500 m from the emitting plant stack. The UV camera was oriented approximately perpendicular to the plume at all three locations. An aperture setting of F-stop 4.5 was used and exposure times ranged from 0.2 to 1.2 s. The images were converted to SO$_2$ pathlength concentration color-maps using an average of calculated calibration cell conversion formulas, and a cross-section of the SO$_2$ concentrations (in ppmm) directly above the stack was recorded (Fig. 3). The similarity of the UV camera retrieval to the plant CEMS data was evaluated in two ways: first, the CEMS data were directly compared to an average of the UV camera SO$_2$ concentrations observed in the cross-section. Secondly, the CEMS data were extrapolated from the sample point concentration (ppm) to a subsequent average pathlength concentration (ppmm) exiting the stack and this value was compared to the UV camera result for average pathlength concentration. These results were also evaluated for changes in accuracy relative to distance from the plume.

3. Results

3.1. Instrumental experiments

The relationship between absorbance and concentration, as calculated for the 14 pressure cell concentrations, is illustrated in Fig. 4. At concentrations less than approximately 1500 ppm, the relationship is linear, with $r^2$ of 0.96. At higher concentrations, the data are best fit with a logarithmic relationship ($r^2$ of 0.914).

Seven images were taken of the pressure cell at a concentration of 814 ppm under challenging weather conditions. The imaging environment varied from high to low ambient light levels and from clear to partly cloudy to snowy background skies. The standard deviation for these measurements is an absorbance of 0.024. If the best-fit line from Fig. 4 is applied to this deviation, it is equivalent to 296 ppm, or 36%.

The consistency of the relationship of absorbance to pathlength concentration of SO$_2$ is addressed in Fig. 5. Each absorbance relationship is determined from images collected at different field sites with varying light quality and at different times of day. The data are derived from images with nearly uniform backgrounds, and with exposure times for each filter image that yielded a maximum DN of >30000 counts. The standard deviations in slope and y-intercept are 1.23×10$^{-4}$ (relative absorbance/ppmm) and an absorbance of −0.004, respectively, indicating the variation from calibration to calibration.

The results of the lens aperture experiments are illustrated in Fig. 6. The vignetting effect in the images is observed in the decrease in illumination towards the corners of the images. The relative illumination values describe the amount of change between the center of the image and the corners, and this varies with the lens aperture. There is a slight but consistent fall-off in DN values at the image corners as the aperture diameter increases from 3.3 mm (F/32) to 23.33 mm (F/4.5), indicating consistent vignetting. The change in illumination is less than 10% for approximately the center half of the image. Analysis of the relationship between illumination value and SO$_2$ pathlength concentration (a proxy for the absorbance relationship) indicates that it remains regular regardless of aperture diameter. Linear fit slopes of calibration cell intensity values (DNs) at seven different apertures had a standard deviation of 1.03DN/ppmm.

3.2. Procedural experiments

Exposure time can vary substantially in the field — a constant value for this parameter can yield dramatically different ranges of DN in images collected under different sky conditions. Consequently, to best illustrate the effect of changing exposure time on images in this experiment, the data are presented with respect to the maximum DN in each image; longer exposure times lead to higher DN (Fig. 7). The relationship between absorbance and SO$_2$ pathlength concentration changes significantly with change in exposure time. The slopes of the best-fit lines increase with length of exposure time, as does the y-intercept (offset due to calibration cell). The rates at which the slopes and y-intercepts change decrease as the maximum image DN approaches the CCD maximum recordable value (65536 counts). The
slopes for images with a maximum DN between 4400 and 7300 change by $2.13 \times 10^{-5}$ and the $y$-intercepts differ by 0.0128, an approximately 20% variation. The exposure times for these images are 0.2 s apart. The changes in slope and $y$-intercept between the images with a maximum DN of 55000 and 65000 counts are only $2.00 \times 10^{-6}$ and $1.20 \times 10^{-3}$, respectively. The exposure times of these images are different by 0.5 s, yet the percent change in slope and $y$-intercept is only about 1%. Images with DN in excess of 32500 counts converge on a single calibration curve.

Fig. 8 gives the results of experiments addressing the procedural variation in optimal or identical exposure times while using the 307 nm and 326 nm filters. Both plots in the figure illustrate six two-filter images (a 307 nm filter subtracting the effect of the 326 nm filter), with each set collected under identical imaging conditions; the only variation is in an exposure time method (optimal or identical). The best-fit lines of the optimal exposures are more consistent in slope and $y$-intercept: the standard deviations for slope and $y$-intercept for optimal exposure are $8.54 \times 10^{-6}$ and 0.005, respectively, while the set taken with the same exposure times has standard deviations for slope and $y$-intercept of $1.47 \times 10^{-5}$ and 0.009, respectively. The average $r^2$ value for the optimal exposure best-fit lines is 0.949, and the average $r^2$ value for the same exposure best-fit lines is 0.767. The optimal exposure method results show a more dependable response for the absorption–concentration relationship, and there is less scatter in the calibration cell data points for each image.

### 3.3. Field testing

During the field testing of the UV camera at the coal-burning power plant, the CEMS data reported SO$_2$ concentrations between 429.1 ppm and 458.2 ppm. The average percent error of four UV camera measurements at both 150 and 1300 m, and an average of three measurements at 1500 m are listed in Table 3. An example of the plume images and resulting comparison to CEMS data is shown in Fig. 9. The heterogeneity of the plume is apparent, illustrating the
from the absolute value of errors. Asterisked results indicate that the average is taken to electrostatic interactions between closely packed molecules or a change in the refractive index (Ingle and Crouch, 1988; Tissue, 1996).

A change in the absorption coefficient results in part from a bias in the spectrum towards longer wavelengths as more SO$_2$ is added to the observation path and the response at more highly absorbing wavelengths becomes saturated. The good fit at lower concentrations ($r^2 = 0.964$) gives confidence in the applicability of the Beer–Lambert law for these calibration cells. Estimated maximum SO$_2$ pathlength concentrations cited in earlier field studies with the UV camera (Mori and Burton, 2006; Bluth et al., 2007) are ≤1200 ppm m, well within the linear relationship observed with our UV camera experiments. Should circumstances arise where the concentrations in the plume exceed the range where the Beer–Lambert law is linear, one option is to derive fluxes from downwind portions of the plume where the transparency of the plume is higher (Andres and Schmid, 2001).

The calibration pressure cell experiments can also be used to evaluate the consistency of a single calibration cell absorption response under changing weather conditions. The pressure cell was imaged repeatedly with a highly variable imaging environment, and the calculated absorbances yielded a standard deviation of 0.024, which is equivalent to 296 ppm/m or 36%. This underlines the dependence of the SO$_2$ retrieval on the observational environment. The relationship between concentration and absorbance may be accurately modeled with each calibration, but an increase in calibration frequency is critical as weather and brightness conditions evolve. The response relationships determined in Fig. 5 again illustrate the variation in the calculated response relationship with variable light conditions. The slopes of the best-fit lines remain relatively constant, indicating that the relationship between absorption at high and low concentrations is regular. However, there is a significant variation in the y-intercept, which is an indication of the offset due to absorption by the calibration cell itself. The relationship is stable but the observed absorption varies with imaging conditions. As demonstrated in Fig. 5, the same value for absorbance can correspond to either 90 or 270 ppm/m depending on the imaging conditions. This demonstrates the importance of regular in situ calibrations.

4.2. Aperture limitation

The results of the aperture experiments indicate that vignetting in the UV camera is affected by aperture diameter, but minimally at the center of the image. The observed darkening effect, after eliminating the possibility of mechanical vignetting from objects in front of the lens, is most likely due to a combination of natural and optical vignetting. The increase in vignetting with aperture indicates optical vignetting, but since it cannot be completely corrected with aperture reduction, it is likely that some vignetting is a property of the lens design (Ray, 2002; van Walree, 2007). This natural vignetting will vary from lens to lens and any other camera configurations (i.e. different lens and/or CCD) would have its own vignetting characteristics. The optical vignetting can be minimized by reducing the lens aperture, but it is important to weigh the subsequent consequence of reduced light levels. This will either result in lower DN values or a choice to lengthen the exposure time, which in turn leads to a lower sample rate and potential smearing of the image. While it is encouraging that the response curve between concentration and absorbance remains constant over a range of aperture settings, the calibration cells were located in the same area of the field of view for each image, and thus can only attest to the minimal variability of the amount of vignetting with change in aperture at the center of the image. Further investigation would quantify the effect of vignetting on the retrieved SO$_2$ pathlength concentration in different regions of the image. For this camera configuration, if we choose to use only areas of the image with less than 20% light reduction from vignetting we will still retain a linear field of view of approximately 900 m at 5 km from the target and 1800 m at 10 km from the target.

4. Discussion

4.1. Calibration cells

The results of the pressure cell trials demonstrate the non-linearity in the absorbance-to-SO$_2$ concentration relationship at concentrations higher than approximately 1500 ppm m. This result is contrary to the linear response expected from the Beer–Lambert law, however, it is not unprecedented. Previous studies (Ayres, 1949; Dick, 1998; Elias et al., 2006) have empirically reported this deviation from the expected response curve at high concentrations of absorbing species, and this may be due to changes in the absorptivity coefficients relating to electrostatic interactions between closely packed molecules or a impracticality of a single pixel comparison to the single point CEMS sample.

Table 3

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Average x-section</th>
<th>Average pathlength</th>
</tr>
</thead>
<tbody>
<tr>
<td>150 m</td>
<td>22.69*</td>
<td>17.81*</td>
</tr>
<tr>
<td>1300 m</td>
<td>−32.21</td>
<td>−38.05</td>
</tr>
<tr>
<td>1500 m</td>
<td>−27.96</td>
<td>−34.17</td>
</tr>
<tr>
<td>Total average</td>
<td>27.59*</td>
<td>29.63*</td>
</tr>
</tbody>
</table>

Percent errors associated with calculated SO$_2$ concentration or pathlength concentration for two comparison methods. Asterisked results indicate that the average is taken from the absolute value of errors.
The method of image processing that utilizes the 307 nm and 326 nm filter images can have the effect of flattening the appearance of vignetting. This method yields a processed image that illustrates only the disparity between absorbing features at 307 nm and 326 nm, and logically would appear flattened as the vignetting effect should be similar for each filter. However, the effect of natural vignetting via the lens design reduces the electronic signal at the corners of the image and, as a consequence, the sensitivity to SO₂ absorption. It may be possible to model the vignetting effect, and thus extrapolate flattened values at the corner of the image, but this region will likely have a decreased signal to noise ratio. This effect is reduced if the DN values in the corners are high, in the same way the signal to noise ratio increases with exposure time, but will occur systematically in all images. Other lenses may cause more or less natural vignetting, but currently the number of off-the-shelf compatible lenses that transmit UV light is small.

4.3. Exposure time

The parameter of exposure time is of critical importance to the UV camera retrieval method, as shown by the results in Fig. 7. The increase in exposure time, leading to higher maximum DN values, essentially increases the signal to noise ratio for each image. The moderation of exposure time for UV camera imaging of plumes is comparable to the function of the automatic gain control (AGC) on a COSPEC instrument (e.g., Stoiber et al., 1983; Andres and Schmid, 2001; Millán, 2008). The current UV camera set-up does not allow for automatic maximization of exposure time (high DN values), and the user must remain vigilant of this parameter at times of rapid change in ambient brightness, e.g., early or late day observations. Our calibration cell response experiments emphasize the benefits of changing the exposure times in calibration images to correspond to changes in plume image exposure times. The results in Fig. 7 indicate that the change in response decreases exponentially with increasing maximum DN and that variability in the response fit approaches 0. For practical purposes, maintaining maximum DN values above 32500 counts will result in a reduction in this error. It is important to note that highly reflective material in an image may lead to DN values that are significantly higher than the rest of the image. These outliers will skew the image DN histogram, and the maximum DN of the image will not refer to the plume or calibration cells. Exposure time adjustments must address the DN range of the target feature.

The increase in signal to noise ratio with lengthened exposure time can be seen in the different results from the “optimal” or “identical” exposure, 307–326 nm filter differing methods. Using the same exposure time for both filters ensures that the maximum DN values for the 326 nm filter image will be large, and those for the 307 nm filter will be significantly smaller. It is particularly degrading to the retrieval results to underexpose the 307 nm filter, as this is the filter tasked with describing the absorption by SO₂. The scatter in the “identical” exposure absorbance values (Fig. 8B) is symptomatic of the increase in noise over signal for the 307 nm filter, and not for the 326 nm filter.

4.4. Accuracy of the technique

The calibration experiment at the coal-burning power plant gives a general insight into the accuracy of the UV camera retrieval technique. The percent errors were within 30% for both the comparison of UV camera SO₂ concentrations to CEMS concentrations and the comparison of UV camera SO₂ pathlength concentrations to the extrapolated CEMS pathlength concentrations. The observations made very near (150 m) to the exit stack were inconsistent in whether they demonstrated an over or underestimation of the CEMS-measured values. The more distant observations using methods 1 and 2 show a consistent underestimation of the CEMS-measured value. An underestimation of the plume pathlength concentration at greater distances is to be expected due to increased UV scattering with distance from the plume. Mori et al. (2006) give evidence of the reduction in measured SO₂ pathlength concentration at distances greater than 1 km due to absorbance attenuation.

The most robust evaluation available for the UV camera retrieval technique is a comparison to measurements directly sampled from a SO₂ plume. However, there are limitations to this calibration technique. In this case, inaccuracies were introduced from timing of the measurements and additional gas species in the plume. The CEMS sample point is located in a duct in the plant, leading to the exit stack. There is a delay between the collection time of this sample and the appearance of this plume parcel above the stack where it is imaged by the UV camera. At the time of this experiment, the UV camera instrumentation included only the 307 nm filter. Thus, another source of error is the presence of absorbing species other than SO₂ in the plume. The use of the 307–326 nm filter differing technique may increase the accuracy of the retrieval. The above sources of error are perpetuated in the second method, where an attempt to extrapolate the CEMS recorded concentrations to a concentration pathlength at the stack exit is performed. For this method, the uncertainty of the exact dimensions of the plume is added as a source of error.

To convert from absorbance to SO₂ pathlength concentration an average of the linear fits observed in the calibration cell experiments.

<table>
<thead>
<tr>
<th>Camera</th>
<th>Observed value (ppm-m)</th>
<th>CEMS Measured value</th>
<th>% Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arg. x-sec. value</td>
<td>471.8</td>
<td>443.4</td>
<td>6.4</td>
</tr>
<tr>
<td>Max. x-sec. value</td>
<td>741.3</td>
<td>443.4</td>
<td>67.2</td>
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<tr>
<td>Extrapolated avg. concentration</td>
<td>471.8</td>
<td>485.19*</td>
<td>-2.8</td>
</tr>
</tbody>
</table>

Fig. 9. A single example of the accuracy assessment using power plant direct sampling data. The image is of an SO₂-emitting coal power-plant stack, taken with the UV camera with the 307 nm filter from a distance of 150 m. The dotted line indicates the approximate location of the measured cross-section.
was used. Calibration response curves calculated from simultaneous or temporally near calibration cell images could increase the retrieval accuracy, and would be the preferable method. However, with the current imaging method constant calibration imaging is not possible, and there exists a trade-off between a continuous record of emissions in the field and frequent calibration images. The results of the power plant experiment are encouraging; the accuracy of the retrieval using even an average calibration response fit is relatively high and indicates that this method has a promising future for field-based plume studies.

5. Summary and future work

These calibration experiments and investigations into the errors associated with the UV camera technique can be used to improve the method for future field campaigns (Table 4). An estimate of the DN value at which the signal to noise ratio begins to plateau (approximately 32,500 counts) suggests a minimum exposure time to be used for field observations. Earlier field methods using a compromised exposure time for the filter differencing technique may not be as accurate as using an optimal exposure time for each filter. The accuracy of the UV camera technique appears to be within the range of current spectrometric methods (10–40% uncertainty [e.g., Stoiber, et al., 1983; Doukas, 2002; Williams-Jones, et al., 2006]), but it is an imperfect comparison for SO2 plume data. The SO2 retrieval is significantly affected by changes in imaging conditions (overall light intensity and uniformity of background). Under these conditions, calibration images should be taken often.

Table 4

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Conclusions</th>
<th>Relevant figures</th>
</tr>
</thead>
<tbody>
<tr>
<td>Instrumental</td>
<td>At concentrations below 1500 ppm m, a consistent, linear relationship fits the concentration vs. absorbance data.</td>
<td>Fig. 4</td>
</tr>
<tr>
<td>Uniformity in image background</td>
<td>The SO2 retrieval is significantly affected by changes in imaging conditions (overall light intensity and uniformity of background). Under these conditions, calibration images should be taken often.</td>
<td></td>
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<tr>
<td>Variability in concentration-absorbance relationship</td>
<td>The degree of absorption is variable with light intensity, but the relationship between high and low concentration absorption is relatively constant. This allows for relative retrievals to be calculated when calibration images are sparse.</td>
<td>Fig. 5</td>
</tr>
<tr>
<td>Image vignetting</td>
<td>Images exhibit regular vignetting which is somewhat dependent on aperture diameter. Vignetting is most likely a result of lens design. Imaging with smaller aperture settings and use of the central 60% of the field of view can dramatically reduce vignetting.</td>
<td>Fig. 6</td>
</tr>
<tr>
<td>Procedural</td>
<td>Exposure time significantly affects accuracy. Exposure time has a significant effect on the retrieval, but the change in the response is dramatically less where exposure times result in maximum DN values greater than 32,500 counts.</td>
<td>Fig. 8</td>
</tr>
<tr>
<td>Double filter method exposure time effects</td>
<td>Using “identical” exposure times for both the 307 and 326 nm filters increases the noise over signal for the retrieval. Using “optimal” exposure times for each filter leads to more consistent calibration relationships.</td>
<td>Fig. 9</td>
</tr>
</tbody>
</table>

Field testing

Power plant calibration
Comparisons show that the UV camera retrieval has an error of no more than ~30%. Table 3

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