Dial Techniques For the Control of Sulfur-dioxide Emissions

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DIAL techniques for the control of sulfur dioxide emissions

Anna-Lena Egeback, Kent A. Fredriksson, and Hans M. Hertz

An extensive program for the evaluation of the DIAL (differential absorption lidar) technique was performed. Measuring and evaluation routines for DIAL measurements on sulfur dioxide in different applications were made. Several field tests were performed with a mobile lidar system employing a Nd:YAG laser pumping a dye laser. Examples of measurements from the work are given. The practical aspects of the technique and the field of applications are discussed.

I. Introduction

Sulfur dioxide is one of the major air pollutants. The main sources are fuel combustion in power plants, combustion for heating and industrial processes, e.g., in smelting plants, paper mills, and sulfuric acid manufacturing industries. The gas is poisonous and may cause serious injuries even at low concentrations. The recommended maximum SO$_2$ concentration in Sweden is 100 $\mu$g/m$^3$ as an average during the wintertime, and the 1-h maximum value is 750 $\mu$g/m$^3$.

In the atmosphere sulfur dioxide combines with water and forms H$_2$SO$_3$ and H$_2$SO$_4$. In the presence of moist air or particles the gas reacts more rapidly and forms sulfates in addition. The reaction products are major constituents of acid rain, which is a large-scale problem in many regions. Acid rain is believed to decrease the growth rate of some forests, especially coniferous woods, and is responsible for the acidification of soil and water, which has led to extermination of fish in many lakes in sensitive regions. The content of mercury in fish, which has led to fishing prohibitions in many regions, is also dependent on the acidification of the lakes. Another effect of the acidification of the underground and freshwater is that the water work system has been damaged and that metals such as copper have precipitated and caused illness to people. Sulfur dioxide is also deposited directly by the vegetation to an extent which is largely unknown.

The problem with acidification is not as severe where the bedrock has a certain lime content, which is the case in many regions. However, in some places, e.g., on the Scandinavian peninsula, the lime content is low and the soil and water cannot neutralize the acid rain and thus the sulfur dioxide emissions have become a serious problem.

During the last few years great efforts have been made to control the sulfur dioxide problem. In many countries the maximum sulfur content allowed in combustible oil has been lowered quite drastically, which has certainly lowered the concentration in urban areas. However, the pollution coming from industrial processes has not been reduced in the same way. One can also fear that there will be an increased emission when coal burning power plants are more frequent, since it is not as easy to control the SO$_2$ from these plants. The situation is now so serious that even unconventional solutions must be considered. Thus, one tries to reduce the acidification of some regions by putting large amounts of lime on the ground and into the water.

There is great need for reliable measurement techniques for the control of SO$_2$ emissions. In some cases, when distinct plumes are the subject of investigation, the emissions can be measured and calculated quite easily with conventional techniques. In most cases however, when several plumes or diffuse emissions are present, it is difficult to determine the flow from a certain area, especially since one must also take the atmospheric influences into account. When coal burning is more commonly used there will be a need for better measurement techniques, since the emissions cannot be calculated in the same way as is possible when burning oil. Since SO$_2$ emission is a large-scale problem there is an increasing need to study the flow and transport of the gas over long distances, which is difficult to do with conventional point monitoring instruments.

There has been an interesting evolution of SO$_2$ monitoring instruments. The possibilities of measuring gas with remote sensing have been improved with the construction of correlation spectroscopy instruments.
and video systems charting the sky spectrum at SO$_2$ UV-absorption wavelengths (e.g., Refs. 1 and 2). These instruments can be very useful in some special applications even if they cannot be used as universal instrumentation to control the SO$_2$ atmospheric pollution. With the employment of artificial light sources in long-path absorption measurements, equivalent to the functioning of laboratory spectrometers, there are possibilities to measure extremely low background concentrations (e.g., Ref. 3).

Laser technology is developed for measuring SO$_2$ using the DIAL technique. In a DIAL measurement one measures the differential absorption of the lidar signal on and off a resonance line of the gas. DIAL and other lidar techniques were reviewed in e.g., Refs. 4–6. During the last few years the DIAL technique has been elaborated for studies on SO$_2$. Some of the early progress was made by Grant and Hake$^7$ and Hoell et al.$^8$ In our group, which was then located at the Chalmers University of Technology, Göteborg, DIAL studies on SO$_2$ were initiated in 1977.$^9$ Laboratory measurements of the absorption spectra of the gas were performed by, e.g., Woods et al.$^{10}$ and Brassington.$^{11}$

When the principles of the technique were demonstrated there still remained an evaluation process of the DIAL technique and the lidar instruments to make measurements reliable and useful to people working on air pollution problems. Different generations of mobile DIAL systems were constructed and tested in field experiments. A Swedish national mobile lidar system was developed in 1979 at the Chalmers University. The construction was based on the results and experiences from the research and previous lidar systems. The mobile laboratory was presented in an extensive paper.$^{12}$ Here only a brief description of the basic design will be given. There are also several mobile lidar systems constructed by other research groups. The Stanford Research Institute,$^{13}$ the research institute of ENEL in Italy, and the National Physical Laboratory in England are working with the DIAL technique and are using mobile systems to study SO$_2$; other groups are starting activities in the field. However, to our knowledge no system has been evaluated and tested in so many different applications and field tests as the Swedish system.

In this work we evaluate the DIAL technique and the mobile system from a practical point of view. We were particularly aiming at the evaluation of effective measurement routines that are useful for solving air pollution problems. During the work we had quite a number of useful contacts with people working with environmental control in industry and the government. We will exemplify the SO$_2$ DIAL technique with a few measurements and discuss measurement strategies which we found useful in the work. Some conclusions will be drawn and plans for future work will be discussed. Applications, in which the technique has proved to be well suited, will also be pointed out. A detailed report on a 2.5-yr period for this evaluation and testing was recently presented.$^{14}$ A paper on the possibilities and the accuracies in DIAL measurements on NO$_2$ was also written.$^{15}$

II. Measurement System

The lidar system platform is a covered truck specially designed for lidar work. Figure 1 is a photo of the system in a typical measurement situation at a paper mill plant. The laser beam is emitted from the system via a large plane mirror in a rotatable dome structure on the roof of the truck. The system has a Newtonian telescope mounted in a fixed vertical position inside the truck, and the backscattered lidar signal is deflected to the telescope by the plane mirror in the dome. The design makes it very easy to steer the laser beam into the desired measurement direction.

The laser system of the mobile laboratory consists of a Nd:YAG laser and a dye laser, which are used in combination when DIAL measurements are made. In the case of SO$_2$ measurements the Nd:YAG laser is pumping the dye laser with the 532-nm output and this second laser is operating with a rhodamine dye. Two frequency doubling crystals are employed for the conversion of the laser light into the UV region. These are tuned to the wavelengths on and off the resonance line, respectively. The dye laser wavelength is changed between each laser pulse and the laser is operating at 10 Hz. The laser power in the UV was only a few millijoules in this work, which was sufficient for measurements at distances up to 2 km. The laser pulse time is $<$10 nsec and the data acquisition system has time windows of 10 nsec, which in practice give a distance resolution of a few meters. The mobile laboratory includes a minicomputer which is used for the steering of the system and also for the calculations and evaluations of the measurements. The entire system is run automatically by the minicomputer during measurements. Great efforts were made to develop measurement and evaluation routines to make reliable studies possible. The system includes gas-absorption cells which are used for a rapid calibration of the dye laser wavelengths. Recently, a hollow-cathode discharge lamp has been included for more precise wavelength calibration. The primary output of the dye laser is then calibrated.

![Fig. 1. Mobile DIAL system in a typical measurement situation at a paper mill.](image-url)
In this part of the UV spectrum, the dye laser, which is operated with a rhodamine dye, in our case rhodamine 640 and rhodamine 610 at wavelengths round 600 nm, and the laser light is frequency doubled to the UV using KD*P crystals.

The background skylight is reduced due to absorption by the atmospheric ozone layers, and the atmospheric extinction of laser light is normally not severe. Further, no interfering absorbing gases exist, with the exception of ozone, which has a comparatively low differential absorption cross section at the studied wavelengths. Another advantage is the well-established laser technique in this part of the UV spectrum. The dye laser is operated with a rhodamine dye, in our case rhodamine 640 or a mixture of rhodamine 640 and rhodamine 610 at wavelengths round 600 nm, and the laser light is frequency doubled to the UV using KD*P crystals which have a high conversion factor. In the work discussed in this paper we used a Nd:YAG laser-pumped dye laser, but in our earlier work we used a flash-lamp-pumped dye laser, which in some cases may be favorable because of its lower cost. However, such a laser system will not give the same distance resolution due to the longer pulse time.

The lidar signal, i.e., the elastically backscattered light from particles and molecules, is measured for two different wavelengths. In this work the SO₂ absorption line at 300.03 nm or the line at 298.0 nm was chosen for the absorption wavelength and the intermediate wavelength 299.3 nm for the reference wavelength. The lidar recordings at the 300.03-nm absorption wavelength and at the reference wavelength are shown in the figure. The evaluation of the measurement is made with the use of the DIAL equation,

\[
\frac{P_{\text{abs}}(R)}{P_{\text{ref}}(R)} = C' \exp \left(-2 \int_{0}^{R} \sigma(\lambda_{\text{abs}}) - \sigma(\lambda_{\text{ref}}) n(r) dr \right),
\]

where \(P(R)\) is the lidar signal at distance \(R\), \(C'\) is a system constant, and \(\sigma\) is the absorption cross section. The factor \(n(r)\) is the concentration of the studied gas at distance \(r\). The quotient in Eq. (1) will decrease at the distances where the gas is present. The calculated DIAL curve is shown at the upper right in Fig. 2. From this DIAL function the concentration of the gas for a given range interval \((R_2 - R_1)\) is evaluated using

\[
n(r) = \frac{\ln \frac{P_{\text{abs}}(R_2)P_{\text{ref}}(R_1)}{P_{\text{abs}}(R_1)P_{\text{ref}}(R_2)}}{2 \int_{R_1}^{R_2} \sigma(\lambda_{\text{abs}}) - \sigma(\lambda_{\text{ref}}) dr}.
\]

An evaluation is shown in Fig. 2, in which a range interval of 50 m was continuously swept with the distance, i.e., the concentration evaluated for a certain distance \(R\) is the mean concentration for the path \(R \pm 25\) m. The appropriate range interval for the calculation is determined by the physical situation. When a plume is studied a range interval of 10 m may be appropriate whereas several hundred meters is to be used when a low background concentration is measured.

The absorption cross-sectional values used for the calculation [Eq. (2)] are critical for accuracy in the measurements. In our calculations we use the cross sections published by Woods et al.\(^{10}\) As pointed out above we do calibrations of the laser wavelengths using a cell containing SO₂ and now in addition employing a hollow-cathode discharge lamp. The gas cell has an SO₂ concentration of 5.4 \(g/m^3\) in pure nitrogen and is kept at atmospheric pressure. From the work done we have a good estimation of the accuracy in DIAL measurements on the gas. A typical DIAL measurement has an accuracy better than 10% and <5 mg/m³ · m. However, the accuracy is very much determined by the weather conditions and other atmospheric parameters. The results given in Fig. 2 are estimated to be good within these limits.

Measurements in one single direction of the type shown in Fig. 2 can in many cases be very useful for the control of the atmosphere in a certain local area. Such
an example is given in Fig. 3, where a set of DIAL measurements in a chemical plant area is shown. The laser beam was directed along a local street at a height of 3–5 m above street level. The concentration was determined in 1-min measurements in the study. Since this was a study where we also wanted to evaluate the accuracy of the mobile laboratory, the absorption wavelength in the DIAL recordings was altered between the absorption lines at 300.03 and 298.0 nm. The distance resolution in these measurements is 50 m. One interesting feature in the study is shown in the figure. In the recording at 10.12 the concentration increased drastically. The increase was due to a sudden diffuse emission of the gas from a sulfuric acid plant. A DIAL system in a case like this is an efficient tool to get immediate information on emission conditions. Measurement time need not be more than a few seconds.

Measurements in different horizontal directions can be performed to chart the SO$_2$ concentration in an area of interest. Such an example is shown in Fig. 4, where DIAL measurements in sixteen different directions were evaluated and put together to form the charting. Each level in the figure represents 100 µg/m$^3$. The measurement area was a paper mill at a distance of ~1 km from the mobile lidar system. Measurements lasting 1 min were made for each direction and thus the time for a scan over the area was 16 min. To get enough statistics and a representative charting of the concentration in the area, nine scans were made during a 2.5-h period. Besides being a demonstration of the technique the measurement results from this field study had a direct impact on the construction of a new treatment system for reduction of SO$_2$ emissions at the plant studied.

Charting of SO$_2$ is a very good application of the DIAL technique, and in practice no alternative method exists to get the same information. Besides using a DIAL system for continuous control and charting of the concentrations in an area, measurements can also be performed for the guidance and relevant localization of point measurement systems in the area. The accuracy in the individual recordings for the charting in Fig. 4 was about the same as in the examples above.

One of the most interesting and important parameters to measure in pollution studies is the flow of a pollutant from an industrial area. In some cases one or a few smokestacks are the dominant sources in an area, but very often the pollution situation is more complex with a large number of smokestacks and perhaps also a considerable diffuse emission, which may be difficult to localize. When only a few smokestacks are present, the pollutants can be recorded with point measuring instruments inside the smokestacks, and the flow of the studied gas can then be calculated. However, one drawback is that there may be difficulty estimating the influences on the flow due to the chemical activities in the smokestacks and in the atmosphere at a close distance, and these influences may be very important. For example, the formation of NO$_2$ from NO in a plume has to be considered when the NO$_2$ flow is to be calculated. When the flow of SO$_2$ is to be estimated the formation of SO$_3$ in the dense plume should be taken into account. When a more complex situation is studied, where many local sources and diffuse emissions are present, the flow is very difficult to calculate and point measuring instruments give limited information.

The DIAL technique offers a very good possibility to study the flow of SO$_2$ from an industrial area. A charting is then made of the gas in a vertical section at
some distance downwind the industry. An example of such a charting is given in Fig. 5, which is a measurement of the SO$_2$ concentration profile in a vertical section downwind a sulfuric acid plant area at a distance of 150 m from the plant. A map of the measurement location is also shown in the figure.

The distributed plume is due to diffuse emissions from the plant, whereas the dense plume at a higher altitude comes from a stack, which is 122 m tall. The readings in the diagram are the integrated total SO$_2$ concentrations in the two plumes in the vertical section. These numbers multiplied with the relevant wind velocity yield the flow of the gas from the area. In this case the wind was measured at an altitude of 15 m using a conventional measurement technique. In addition to this the wind was measured at the 70-m level by studying the velocity of a particle plume. The plume velocity was calculated with good accuracy using an ordinary digital watch and by measuring the distance and wind direction with the lidar system. In the example given the total flow of SO$_2$ was found to be 140 g/sec or 500 kg/h. The accuracy in the study is limited by the difficulty of estimating the wind velocity at different altitudes from measurements at only two altitudes. In this individual measurement the flow is expected to be good within 30%, whereas the average flow could be determined much better in the study. The diffuse emission from the plant studied was found to vary more than 1 order of magnitude, whereas the dense plume was emitted by a very stable source. The example given here shows the usefulness of the DIAL technique for measuring total flows. Clearly, the chartings can also be used to distinguish different sources.

The form of presentation of measurement results is very important in all air pollution studies. Very often the amount of measurement data is very large and the individual results from different points in the studied region and at different times are of no relevance but must be put together to be of any value for the study. The DIAL technique is quite favorable in this respect when, e.g., the spatial distribution of the SO$_2$ pollutant is to be determined. The measurements are normally evaluated in terms of concentrations along the measurement direction as shown in Fig. 2. A set of recordings in different directions can then be put together to form a charting of the concentration representative for a certain time interval, which was exemplified in Figs. 4 and 5. In this work the evaluation of DIAL measurements was computerized and could be done rapidly. The actual drawing of the chartings was made by hand and took some time. There is no problem computerizing the drawing, and routines for this are being developed. Figure 6 is another example of charting a plume in a vertical section, in a form of presentation which displays the individual measurement results.

The diagram is not as perspicuous as the former examples, but it describes the measurement results in a more direct way. A condition for the chartings to be drawn in the former examples is that the atmospheric situation be relatively stable, since the different vertical directions are not studied simultaneously. It would hardly be meaningful to draw concentration contours in Fig. 6 as the atmospheric situation apparently changed during the measurement. In this case the alternative form of presentation describes the measurement result better, giving the actual result for each direction without any spatial smoothing. The best way
of presenting measurement results like these would certainly be to use a color display. The measurement in Fig. 6 was made during the sixth CEC Joint Campaign on Remote Sensing of Atmospheric Pollution in France in June 1983. The plume studied was emitted from an oil refinery.

As was pointed out in the discussion on the horizontal charting of concentrations in Fig. 4, some measurement time is needed to make a representative charting for a certain area because of the fluctuating atmospheric conditions. The same holds for a vertical charting, and the measurement times for the chartings in Figs. 5 and 6 were 50 and 20 min, respectively. However, in many cases it is sufficient to determine the total flow of the gas from an industrial area, i.e., detailed information on the distribution in the vertical section is not needed. Then there is the possibility of doing rapid measurements by continuously scanning the laser beam during the measurement. Technically this means that, instead of storing the measurement data in a large number of data files, the data are stored in only a few files and therefore the computer time is very much reduced. In principle a rapid charting of the type shown in Figs. 5 and 6 would also be possible, but then a very large multichannel memory would be needed to handle and store the data. Another form for presentation of a total SO₂ flow, which is useful also in rapid measurements, is given in Fig. 7. The horizontal direction for the vertical section is leeward of a large metallurgical plant. This form for presentation and calculation of measurements has turned out to be very useful in our studies. The curves in Fig. 7, which are evaluated from eighteen measurements, display the integrated SO₂ content in g/m for different vertical subsections. The total flow was estimated to 670 g/sec or 2400 kg/h. The measurement results were weighted with the appropriate wind velocity in the calculation. The wind velocity was calculated for different altitudes with the technique discussed in the former example. The accuracy in this flow measurement was estimated to be within 30% and it is mainly limited by the uncertainties in the wind velocity determinations.

Figure 8 shows another measurement example in a more perspicuous form of representation. The measurement was performed at the same location as the measurement shown in Fig. 5. The SO₂ concentration integrated with the altitude, i.e., the horizontal projection of the plume, is displayed. The content in a dense plume and the diffuse emissions from sulfuric acid plants are distinguished in this recording. The readings in the figure are the integrated contents of SO₂ in the section. The corresponding flows were calculated to 120 and 50 g/sec, respectively. The wind data were obtained in the same way as in the measurement examples above.

Figure 9 is still another way of displaying measurement results from a flow study. The measurement was made on a spreading plume from a heating plant. The horizontal projection of the plume is displayed as in Fig. 8 and in addition a vertical projection is given in an approximate altitude scale. Since the vertical direction was changed stepwise in the measurement the projection is displayed as a step function.
emissions in industrial areas is an obvious application
of use to study the transport of pollutants across the
plant. This form of presentation has turned out to be well suited for
rapid evaluations in field work and is also adapted to plume dispersion
models.

IV. Conclusions

DIAL has become a useful measuring technique for
applications in air pollution studies, not only in dedi-
cated campaigns but also in routine work to control air
quality and pollution emissions. In this work quite a
few field studies were made and the technique was
tested in realistic cases where all kinds of practical
problem were encountered. A few typical examples on
measurement results are given in this paper. The work
showed that a DIAL system, which includes such deli-
crate equipment as advanced Nd:YAG and dye lasers,
delicate optics of high quality, and a minicomputer with
floppy disks, can be put together in a small truck and
be transported on bumpy roads and still be effectively
operated in an industrial environment. The need for
this remote sensing technique to measure SO$_2$ was evi-
denced in one of the field tests, in which the measure-
ment results from a DIAL charting led to the con-
struction of a new treatment system for reduction of the
emissions at the studied plant. Another expressed
evidence is that an industry, which is one of the major
pollutant sources of SO$_2$ in Sweden, has made a decision
for such measurements to control the flow from an in-

The sphere of applications of the DIAL technique for
SO$_2$ measurements is exemplified in this work, but there
are also other areas where the technique can be useful.
The monitoring and charting of diffuse and source
emissions in industrial areas is an obvious application of
the technique. Studies of the spreading of gas from
a source and its effects on the air quality in surrounding
areas are also important. Measurement of total flow
is one of the most interesting applications. The need
for such measurements to control the flow from an in-
dustrial area is evident, but the technique might also be of
use to study the transport of pollutants across the
borders. Not least, DIAL is a remote measuring tech-
nique for research on air pollution problems, e.g., to
determine the washout of SO$_2$ by rain and to evaluate
models for the spreading of pollutants. The extent of
DIAL activities will primarily depend on the interest
and decisions from industry and authorities, whereas
the potential tasks are many and the technique has been
shown to work in practice.

The measurement accuracies in SO$_2$ DIAL studies
were investigated and a few examples are given in this
paper. The accuracies are partly determined by the
lidar system parameters which are further discussed in
Ref. 15. In practical work the signal-to-noise ratio in
a measurement is limited also by weather conditions
and other atmospheric parameters, e.g., the presence
of diffuse particle plumes. Therefore it is difficult to
give general numbers for the accuracies in DIAL mea-

Fig. 9. Vertical and horizontal projections of a plume from a heating
plant. This form of presentation has turned out to be well suited for
rapid evaluations in field work and is also adapted to plume dispersion
models.

In the evaluation project for the mobile system, rou-
tines for measurements on NO$_2$ and O$_3$ were also tested.
The continuing research includes the extension of lidar

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References


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July


23–27 3rd Int. Conf. on Infrared Physics, Zurich. F. Kneubuhl, Solid State Physics Lab., ETN Moenggberg, CH-8093 Zurich, Switzerland

23–27 Photographic Science course, Rochester. B. Reinherr, RIT, P.O. Box 9887, Rochester, N. Y. 14623

August

1–3 Int. Conf. on the Physics of VLSI, Palo Alto. K. Saraswat, Stanford Electronics Lab., Stanford U., Stanford, Calif. 94305

5–9 Rocky Mountain Conf., Denver. J. Gurnsey, 5531 Bitterbush Way, Loveland, Colo. 80537


6–10 Tunable Lasers course, Boston. Laser Inst. of Amer., 5151 Monroe St., Suite 118W, Toledo, Ohio 43623

12–17 2nd Int. Symp. on Diffuse Reflectance Spectroscopy, Chambersburg, Pa. P. Williams, Grain Res. Lab., 1404-303 Main St., Winnipeg, Manitoba R2C 3S8, Canada


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