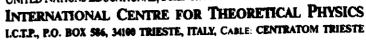


INTERNATIONAL ATOMIC ENERGY AGENCY UNITED NATIONAL CENTRE FOR THEORETICAL PHYSICS







UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION



INTERNATIONAL CENTRE FOR SCIENCE AND HIGH TECHNOLOGY OF INTERNATIONAL COURSE FOR TRIBORAL COURSE FOR TRIBO

SMR/760-37

"College on Atmospheric Boundary Layer and Air Pollution Modelling" 16 May - 3 June 1994

"Use of a Mobile UV-Dial System for SO_2 Monitoring"

A. MARZORATI ENEL Direzione Studi e Ricerche - CRAM Milan, Italy

Please note: These notes are intended for internal distribution only.

USE OF A MOBILE UV-DIAL SYSTEM FOR SO₂ MONITORING

A. Marzorati and G. Elisei

ENEL / DSR (Research and Development Direction)
CRTN (Thermal and Nuclear Research Centre) - Milan (Italy)

Abstract

ENEL (Italian Electricity Generating Board) is interested in the knowledge of transport and diffusion properties of SO₂ and other pollutants in the troposphere connected with the production of power energy.

To this purpose, a mobile UV-DIAL system for SO₂ monitoring has been developed. It uses a dot bled dye laser pumped by the second harmonic of a Nd-YAG laser with 20 Hz repetition rate. Fully automatic measurements can be made by means of a computer-controlled electronic device.

Since 1982, this system has been used during experimental campaigns in different places (industrial and urban sites) to track plumes near the power plants, monitor SO₂ over large zones and also to measure gas emissions from soil and fumaroles in volcanic areas.

The main results obtained during some of these surveys will be also presented.

1. Introduction

Historically air pollution monitoring has been performed using chemical techniques and sampling for later laboratory analysis. Actually LIDAR (LIght Detection And Ranging) and DIAL (Differential Absorption LIDAR) techniques, using lasers, can provide tridimensional, in-situ, in real-time and non intrusive pollution concentration measurements.

These methods can provide the following types of measurements:

- monitoring of the pollutant sources and concentrations over very large areas with high spatial resolution;
- measurement of the pollutants transport for diffusion models validation;
- monitoring of gases important for atmospheric photochemistry and physical atmospheric parameters like temperature and wind velocity;
- monitoring leakage from chemical plants and pipelines.

In this paper the principle of the LIDAR/DIAL technique is briefly discussed with particular regard to a short description of ENEL UV-DIAL system. Its calibration and intercomparison with other similar or different devices is also presented. A great importance is given to the use of the DIAL system in various fields of application with different objectives - industrial areas (plume tracking, SO₂ ground level concentrations, down-wash effect), urban areas (SO₂ vertical profiles), volcanic areas (SO₂ flux measurements).

2. Principle of the LIDAR/DIAL technique

A LIDAR uses the backscattered signal of a laser beam from the atmosphere to make measurements of its properties by means of scattering and absorption. It is generally used in a monostatic configuration, where the transmitter and the receiver are located in the same point. The use of laser sources allows many advantages for atmospheric diagnostics, because laser radiation can have a very narrow spectral width, the wavelength can be tunable and laser intensities can be generated in very short pulses. This last property is particularly useful because it can provide range informations.

The LIDAR signal backscattered by the atmosphere, at a given wavelength, can be determined by the formula (Collis, 1970):

$$W(R) = Q \cdot W_1 \cdot \frac{c \tau}{2} \cdot \frac{A}{R^2} \cdot \beta(R) \cdot \exp \left(-2 \int_0^R \alpha(r) dr \right)$$
 (1)

where:

W(R) is the optical power received from distance R;

Q is the telescope optics efficiency;

W_t is the laser transmitted power;

c is the light speed;

 τ is the laser pulse duration;

A is the telescope area;

 $\beta(R)$ is the backscattering coefficient at distance R;

 $\alpha(R)$ is the extinction coefficient at distance R.

The term $\beta(R)$ is the sum of Raleygh and Mie backscattering coefficients, while $\alpha(R)$ is given by the formula:

$$\alpha(R) = \alpha_R(R) + \alpha_M(R) + \alpha_{GAS} \qquad (2)$$

where $\alpha_R(R)$ and $\alpha_M(R)$ are respectively the Rayleigh and Mie extinction coefficients and α_{GAS} is equal to K·P(R) (K is the gas absorption coefficient and P(R) its partial pressure at distance R).

In the DIAL method, a tunable two wavelengths laser is required. One wavelength (λ_{OR}) is tuned to match an absorption line peak of the researched gas, while the second wavelength (λ_{Off}) is tuned to a region of low absorption (see, for instance, the SO₂ absorption spectrum in Fig. 1).

Comparison of the two signals gives a direct concentration measurement of the pollutant gas. If λ_{OR} and λ_{Off} are close enough, the variations of Rayleigh and Mie coefficients can be neglected. The following expression:

$$P(R;R+L) = \ln \left[\frac{W_{on}(R+L) \cdot W_{off}(R)}{W_{on}(R) \cdot W_{off}(R+L)} \right] \cdot \left[\frac{1}{-2 \cdot (K_{on} \cdot K_{off}) \cdot L} \right]$$
(3)

gives the average partial pressure of the gas along the path from R to R+L, where W_{on} (W_{off}) and K_{on} (K_{off}) are respectively the received powers and gas absorption coefficients at the wavelengths λ_{on} (λ_{off}). L is connected with the DIAL system spatial resolution and it is limited by the laser pulse length and receiver electronic bandwidth. In our case, with 100 nanoseconds sampling time for the transient recorder, a 15 meters minimum spatial resolution is achieved.

3. Description of ENEL UV-DIAL system

The UV-DIAL system utilizes a doubled dye laser pumped by the second harmonic of a Nd-YAG laser with 20 Hz repetition rate. The emission at $\lambda_{\rm on}$ and $\lambda_{\rm off}$ is changed shot by shot by means of a device that inserts a prism in the dye laser oscillator cavity; in this way, the laser beam is deflected on the grating of the laser cavity that selects the wavelength. The two wavelengths utilized for SO₂ measurements are shown in Fig. 1.

The laser beam is coaxially transmitted by means of steering optics to the telescope. This can rotate over a solid angle of 2π with a precision of 0.1° in zenith and 0.25° in azimuth.

The optical signal is detected by a photomultiplier, whose gain is modulated in time with a quadratic law. In this way, the dynamic of the DIAL signal is greatly reduced with improvement of precision in A/D conversion. The digitazed signals are then transmitted to a DEC VAXStation GPX/II.

Through the electronic control and timing system, the computer is able to make fully automatic measurements. It is possible to program telescope rotation, laser firing, wavelengths change and other functions. The software permits measurements over a fan of directions, with real-time data processing. The results, that are SO₂ concentrations as a function of distance, are stored on hard disk and subsequently can be recalled to get concentration maps using a printer-plotter. Table 1 summarizes the system specifications.

Table 1 - ENEL UV-DIAL characteristics

TRANSMITTER

LASER SYSTEM: Nd-YAG pumped dye laser

ENERGY OUTPUT : 300 mJ (at 530 nm)

10 mJ (at 300 nm) 10 mJ (at 490 nm)

30 mJ (at 720 nm)

LASER BANDWIDTH : 0.1 cm⁻¹
REPETITION RATE : 20 Hz
PULSE DURATION : 10 ns

RECEIVER

TELESCOPE AREA : 0.25 m²
OPTICS EFFICIENCY : 20% (SO₂)
FIELD OF VIEW : 1 mrad

DETECTOR : Photomultiplier PHILIPS XP-2020Q

DETECTOR SPECIFICATIONS : 20% quantum efficiency
TRANSIENT RECORDER : SONY TEKTRONIX 390 AD

(10 bit, 10MHz)

PERFORMANCES

RANGE : up to 3 Km
SENSIBILITY : 50 ppb (SO₂)
SPATIAL RESOLUTION : 15 to 200 m
TELESCOPE ANGLE RESOLUTION : 0.1° in zenith

0.25° in azimuth

The system, installed on a truck and equipped with a power generator of 50 KVA, is provided with remote control of laser beam alignment. Also a TV camera is mounted on the telescope for eye safety and pointing control.

The total time taken for the measurement in one direction, including telescope movement, is about 10-30 seconds; the set-up time for the whole system is around 2 hours.

During the 6th EEC Campaign on Remote Sensing at Fos-Berre in June 1983, it was demonstrated that DIAL systems can play an important role in measuring atmospheric pollutant dispersion (Capitini et al., 1984; Marzorati et al., 1984) and in testing mathematical models over complex terrain or near land-water interfaces.

An intercalibration was performed on June 3rd and 4th, 1983 among the DIAL systems taking part in the campaign, i.e. NPL (UK), Lund (Sweden), CEA/EERM (France) and ENEL (Italy), on the hill overlooking Martigues.

For the purpose of calibration, the four DIAL systems (placed at distances of less than 100 meters) were pointed in the same direction. Measurements were taken over the same period of time, with identical ranging patterns (500-1000 meters, 1000-1500 meters, etc.).

Fig. 2 shows the comparison obtained in the morning of June 3rd, 1983 and in the afternoon of June 4th, 1983 in the 500-1000 meters range between CEA/EERM and ENEL DIAL systems. Notwithstanding the positions of the two systems were not exactly the same, there was a good agreement in the results.

4. Experimental results

In order to fulfil the requirements of every kind of campaign, the software was developed to comply with the following requirements:

- only one type of data processing to satisfy the different experimental situations;
- real-time data concentration data results;
- estimate of experimental error on concentration measurement linked with its value.

Table 2
Experimental campaigns performed during the period 1982-1989

TYPE OF MEASUREMENT	HOURS OF WORK
Power Plants Plume Tracking (for different aims) MONFALCONE SANTA GILLA	1500
Refineries FOS-BERRE EEC 1983 CAMPAIGN	200
Industrial Areas SULCIS, <u>VADO LIGURE</u> FOS-BERRE EEC 1983 CAMPAIGN	700
Urban Areas ROME	400
Other (Volcanic, Geothermal Areas) VULCANO ISLAND	200

Since 1982, the system has been used during experimental campaigns in different sites (industrial or urban) to track plumes near the power plants, to monitor SO₂ over large areas (Marzorati et al., 1984) and also to measure gas emissions from soil and furnaroles in volcanic areas (Corio et al., 1984; Carapezza et al., 1989). Table 2 summarizes the main experimental campaigns performed in the period 1982-1989 (in particular, the surveys to which this paper refers further are underlined).

4.1 Power Plants Plume Tracking

This problem is very important in sites characterized by a complex orography and/or by a breeze regime, where wind vertical profiles (speed and direction) can not be easily foreseen. As a matter of fact, the plumes path may be strongly influenced by wind speed and direction at heights higher than the stacks ones, where standard instrumentation (for example, cup anemometers) is generally located.

Plumes trajectories can be forecast by using devices like Doppler Sodars and R.A.S.S. (Brusasca et al., 1986), but the use of a DIAL system gives the possibility to check them even when multiple sources are present.

An experimental campaign with the use of UV-DIAL system was performed in Monfalcone area during June 1985 with the purposes to measure SO₂ in the plumes emitted by the power plant stacks, to determine the prevalent directions of pollution transport with height and also to monitor SO₂ almost at ground level to verify the accuracy of chemical monitoring network.

The DIAL system was located at a distance of about 800 meters in direction N-NW from the power plant; the possible plumes path was estimated by pibal soundings and vertical scans in various directions were performed to measure plumes pattern and SO₂ concentration (every single measurement is obtained by averaging 200 laser shots with a spatial resolution of 15 meters at different elevation angles).

In Fig. 3 it is represented the plume vertical section and its horizontal dimension obtained on June 19th, 1985 from 6:22 a.m. to 7:35 a.m., while in Fig. 4.a and 4.b the measurements performed at 6:22 a.m. and 6:57 a.m. respectively in direction 185° N and 215° N are shown.

From Fig. 3, it can be derived that the plume rise was about 370 meters (at a distance of approximately 1500 meters from the power plant stacks); in this situation, very low SO₂ g.l.c. were measured by the chemical monitoring network. An estimate of the distance where you should have the SO₂ maximum g.l.c. can be done by using the effective height H_e derived from the previous measurements. In our case, an effective height H_e equal to 520 meters shows that the maximum SO₂ g.l.c. was probably at a distance of about 2 Km from the power plant (the Briggs "urban" scheme for C category was used).

In some situations it is also important to verify if plumes are trapped in layers interested by down-wash effects of buildings or low stacks, especially in the neighbourhood of very peopled towns.

This phenomenon can be studied by using water channels or wind tunnels, but some errors are introduced in the results when related to real dimensions, even if their accuracy can be considered quite good.

During an experimental campaign performed in January 1984 close to Santa Gilla power plant (a small installation with low stacks in Cagliari area), the DIAL system was used not only to track plumes, but also to estimate the critical wind speed for which plumes were trapped by down-wash effects.

The persistent wind direction, measured with a cup anemometer at 10 meters height, allowed repeated measurements in vertical sections at distances between 60 and 1000 metres downwind to the power plant (every single scan is obtained by averaging 200 shots with a spatial resolution of 15 meters at different elevation angles). In Fig. 5.a and 5.b some vertical scans in different directions are shown, while in Fig. 6 it is represented the link between wind speed and distance from sources where the plumes were bending down to a 30 meters height (at lower heights, the measurements were not possible for safety reasons).

The experimental results show that the downfall distance could be estimated with a better accuracy interpolating the "squares" - in Fig. 6 it is represented by a dashed line. Referring to Fig. 6 again, it can be seen that there was a first down-wash effect with 4 m/sec wind speed, while a wind speed higher than 6 m/sec could produce a continous plume downfall (it must be taken into account that the diagram in Fig. 6 refers to this specific situation with low stacks - 36 meters and 42 meters high - and buildings approximately of the same height - 30 meters and 25 meters high).

4.2 Industrial Areas

One of the most interesting results was obtained during an experimental campaign performed in March 1987 along a complex shoreline on the Ligurian Sea close to Vado Ligure power plant and other industrial installations. The main purpose of this survey was to characterize pollution episodes due to winds blowing towards the sea. A map of Vado Ligure area is shown in Fig. 7: the DIAL system was located on a wharf at a distance of about 1.2 kilometers E from the power plant and industrial area.

The measurements were performed at two different elevation angles (7° and 14°), scanning the telescope over a fan of 21 directions (180° N - 330° N) and averaging 1000 shots with a spatial resolution of 120 meters. The first measurement - at 7° elevation angle - can characterize atmospheric layers in which low emission sources generally diffuse (at a distance of 2 Km from the DIAL position, the relative height is about 250 meters); on the contrary, the second one - at 14° elevation angle - is representative of that part of Planetary Boundary Layer (PBL) interested to the diffusion of

plumes emitted from Vado Ligure power plant (at a distance of 2 Km from the DIAL position, the relative height is about 500 meters) [see Fig. 8].

Fig. 9.a and 9.b show two horizontal scans performed on March 9th, 1987: in the first measurement (Fig. 9.a), SO₂ values due to low emissions were evaluated, while in the second one (Fig. 9.b) the Vado Ligure power plant plume pattern was easily detected.

4.3 Urban Areas

During January 1987, an experimental campaign was performed in Rome with the DIAL system located on one of its Seven Hills - Gianicolo - at a height of about 100 meters. The measurements, averaged on 200 shots with 15 meters spatial resolution, were taken in the same way described for Vado Ligure campaign in paragraph 4.2 (see Fig. 8), but at elevation angles of 1.5° and 6.5° (they were the minimum angles that allowed not to have obstacles along the optical path). Scans at higher elevation angles were sometimes done with the purpose to eventually verify diffusion phenomena in higher atmospheric layers.

Every set of measurements could give both SO₂ vertical and radial profiles through an appropriate computation: in Fig. 10, the results obtained on January 14th, 1987 from 5:45 a.m. to 4:44 p.m. are presented. The first diagram (on the left) represents SO₂ vertical profile: every asterisk is the average of available data at the same height on different directions. The dashed line connects the SO₂ averages of every height, while the continuous line is the SO₂ vertical gradient regression curve. On the right side of Fig. 10, three diagrams represent the radial SO₂ concentration profiles along the optical path at different elevation angles. The shape of the diagrams is quite regular with SO₂ concentration values around 60 μ g/m³, typical data during a disturbed meteorological situation with strong winds and intermittent rain.

4.4 Volcanic Areas

The study of natural inputs of sulphur and carbon compounds can provide the basis for a reliable estimate of the anthropic to natural emissions ratio, which represents the reference point in order to evaluate the man induced alteration of geochemical cycles, on global as well as on a regional basis. The study of the pathways of circulation and fate of these compounds naturally emitted can, moreover, get reliable information in order to predict the circulation and fate of the selfsame compounds put into the environment by human activities, and viceversa.

An equally important aspect refers to the volcanic risk surveillance. It is well established that the relevant variations in the sulphur chemical species flow patterns occur before volcanic eruptions and/or explosions. Their trend, with the real-time control, can give an useful aid in volcanic surveillance.

The results achieved in the Vulcano Island surveys during November 1983 and 1989 point out that the SO₂ plumes from a small volcano were easily detected and measured. In the first campaign (Carapezza et al., 1989), it proved difficult to obtain reliable SO₂ flux estimates, also because of atmospheric turbolence downwind from the volcano cone, which was the only zone where it was possible to detect the plumes. In the second one, fine meteorological conditions and a better location of the DIAL system allowed to have good measurements (see, for instance, Fig. 11.a and Fig. 11.b) and reasonable SO₂ flux estimates, especially with N-NW winds as on November 14th, 1989 (Fig. 12). Unfortunately a comparison of these results with the previous ones was not possible for the reasons explained before, but DIAL SO₂ flux estimates are in good agreement with those ones measured at the same time with other remote sensing instruments (i.e. COSPEC), that have shown a significant increase of volcanic activity since 1983.

The results achieved in these surveys indicate that DIAL techniques can be also advantageously applied to the study of natural systems apart from the control of man induced pollution.

5. Conclusions

The DIAL method has demonstrated its capability to perform remote atmospheric analyses and measurements, which are hardly possible with conventional tools. The experimental surveys described in this paper show that the DIAL system can supply data for diffusion models over large regions, but it is also a very important device for environmental protection.

DIAL ability to discriminate emissions with good detail is very important not only in complex terrain and with different distributed sources, but these systems can give a powerful aid in volcanic risk surveillance due to the circumstance that they can get real-time information on SO₂ flow pattern, as premonitory events of a volcanic eruption or explosion.

Some improvements of this UV-DIAL system (change of the dye laser with a solid state source - Ti:Al₂O₃) will give the possibility to measure other chemical compounds (O₃, NO, NO₂) and to extend the measurement range to 5-6 kilometers due to output laser energy increase (20 mJ for SO₂). Moreover, a NIR-DIAL system for measurement of water vapour, pressure and temperature and a Wind Doppler LIDAR are also in further development.

Acknowledgements

The authors are very grateful to Mr. G. Bocchiola (ENEL/CRTN) for his technical contribution during the experimental campaigns and helpful cooperation in data analysis.

References

- Brusasca, G., Marzorati, A. and G. Tinarelli (1986). Software for Real-Time Acoustic Remote Sensing Instrumentation and Atmospheric Pollution Control. Proc. Int. Conf. ENVIRONSOFT 86. pp. 759-769, Los Angeles, CA, November 1986.
- Capitini, R., Ancellet, G., Megie, G., Pelon, J. and D. Renaut (1984). DIAL LIDAR Measurements of Atmospheric Pollutants (SO₂, O₃) during the Fos-Berre 83 Experiment. <u>Proc. 12th Intern. Laser Radar Conf.</u> pp. 269-273, Aix-en-Provence, August 1984.
- Carapezza, M., Dall'Aglio, M., Falchi, G. A., Elisei, G., Marzorati, A. and E. Zanzottera (1989). Measurements of Volcanic SO₂ in the Atmosphere by DIAL Remote Sensing Techniques Results Obtained in the Vulcano Island Survey. Proc. 8th World Clean Air Congress, "Man and His Ecosystem", 3, pp. 77-82, The Hague, September 1989.
- Collis, R.T.H. (1970). LIDAR, Applied Optics, 9, 1782.
- Corio, W., Draghi, S., Ferrario, A. and E. Zanzottera (1984). LIDAR Applications to the Geochemical Field. <u>Proc. CLEO</u> 84, pp. 160-163, Anaheim, CA, June 1984.
- Marzorati, A., Corio, W. and E. Zanzottera (1984). Remote Sensing of SO₂ during Field Tests at Fos-Berre in June 1983. <u>Proc. 12th Intern. Laser Radar Conf.</u>, pp. 259-262, Aix-en-Provence, August 1984.

Figures captions

- Fig. 1 SO₂ absorption spectrum.
- Fig. 2 SO₂ integrated concentration in the range 500-1000 meters from DIAL location (CEA/EERM and ENEL).
- Fig. 3 Plumes vertical section and its horizontal dimension obtained during Monfalcone campaign on June 19th, 1985 from 6:22 a.m. to 7:35 a.m..
- Fig. 4 a) SO₂ plume pattern obtained during Monfalcone campaign on June 19th, 1985 at 6:22 a.m. in direction 185° N.
 - b) As in Fig. 4.a, but at 6:57 a.m. in direction 215° N.
- Fig. 5 a) SO₂ plume pattern obtained during Santa Gilla campaign on January 9th, 1984 at 3:13 p.m., 3:26 p.m. and 3:40 p.m. respectively in direction 41.5° N, 45.5° N and 55.5° N.
 - b) As in Fig. 5.a, but at 3:52 p.m., 4:08 p.m. and 4:19 p.m. respectively in direction 69.5° N, 81.5° N and 93.5° N.
- Fig. 6 Relationship between wind speed and distance from sources where the plumes were bending down to a 30 meters height (the circles and squares represent respectively the first and second measured plumes downfall).

- Fig. 7 Map of Vado Ligure area the DIAL system, which position is represented with a cross, was located on a wharf at a distance of about 1.2 kilometers E from the power plant and industrial area.
- Fig. 8 Relative heights (referred to DIAL position) at 7° and 14° elevation angles during Vado Ligure campaign.
- Fig. 9 a) SO₂ plume pattern obtained during Vado Ligure campaign on March 9th, 1987 at 7° elevation angle in direction 180° N 330° from 7:56 p.m. to 8:45 p.m..
 - b) As in Fig. 9.a, but at 14° elevation angle from 8:46 p.m. to 9:34 p.m..
- Fig. 10 SO₂ vertical and radial profiles obtained during Rome campaign on January 14th, 1987 from 5:45 a.m. to 4:44 p.m..
- Fig.11 a) SO₂ isoconcentration map obtained on November 14th (6:33 p.m.-6:37 p.m.) by a vertical scan in direction 130° N, averaging 200 shots with a spatial resolution of 15 meters. The section is approximately 500 meters far from the volcano crater (wind speed = 3.5 m/sec; wind direction = N).
 - b) SO₂ isoconcentration map obtained on November 16th (9:41 a.m.-9:55 a.m.) by a horizontal scan at 9° elevation angle, averaging 200 shots with a spatial resolution of 15 meters (wind speed = 3.5 m/sec; wind direction = E).
- Fig. 12 SO₂ flux estimates during 2nd Vulcano Island survey on November 14th, 1989 (the fluxes are given in tons/day).

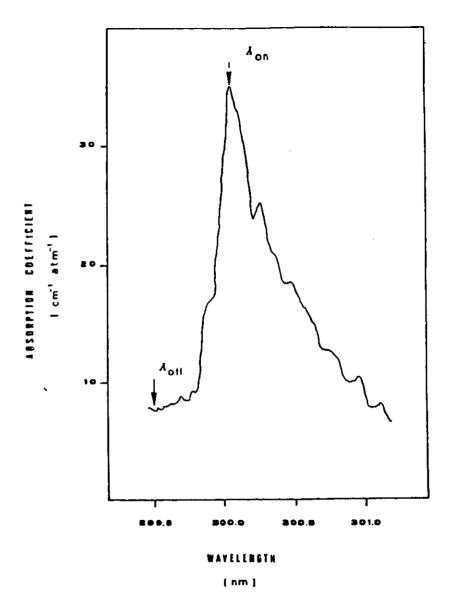
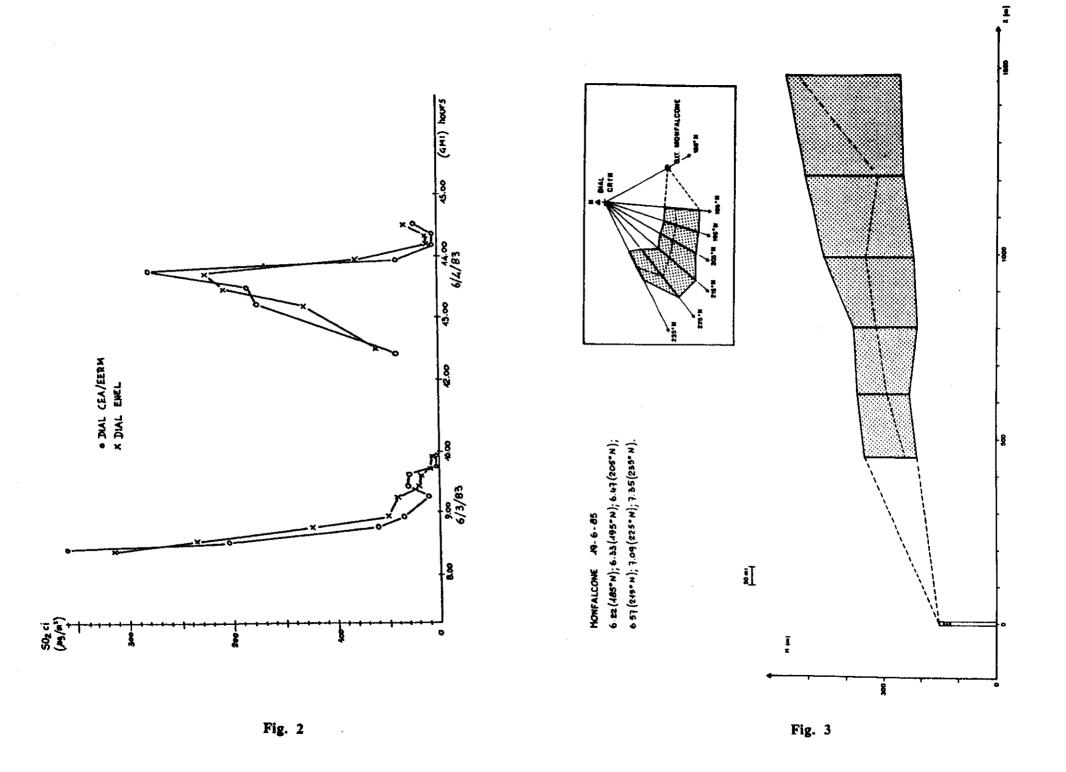


Fig. 1



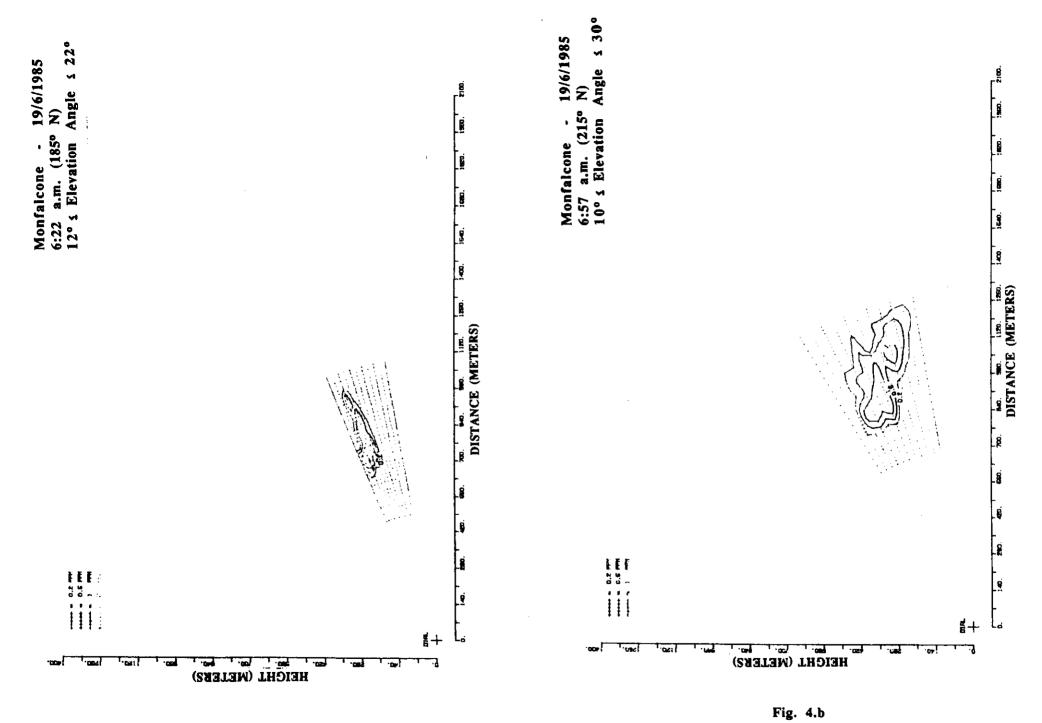


Fig. 4.a

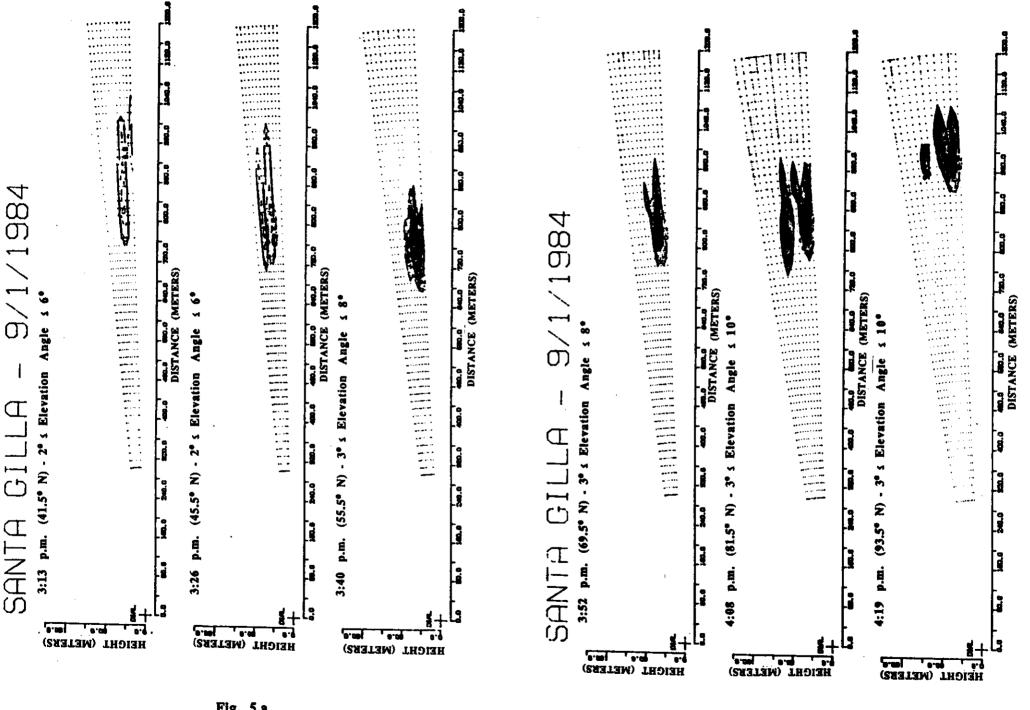
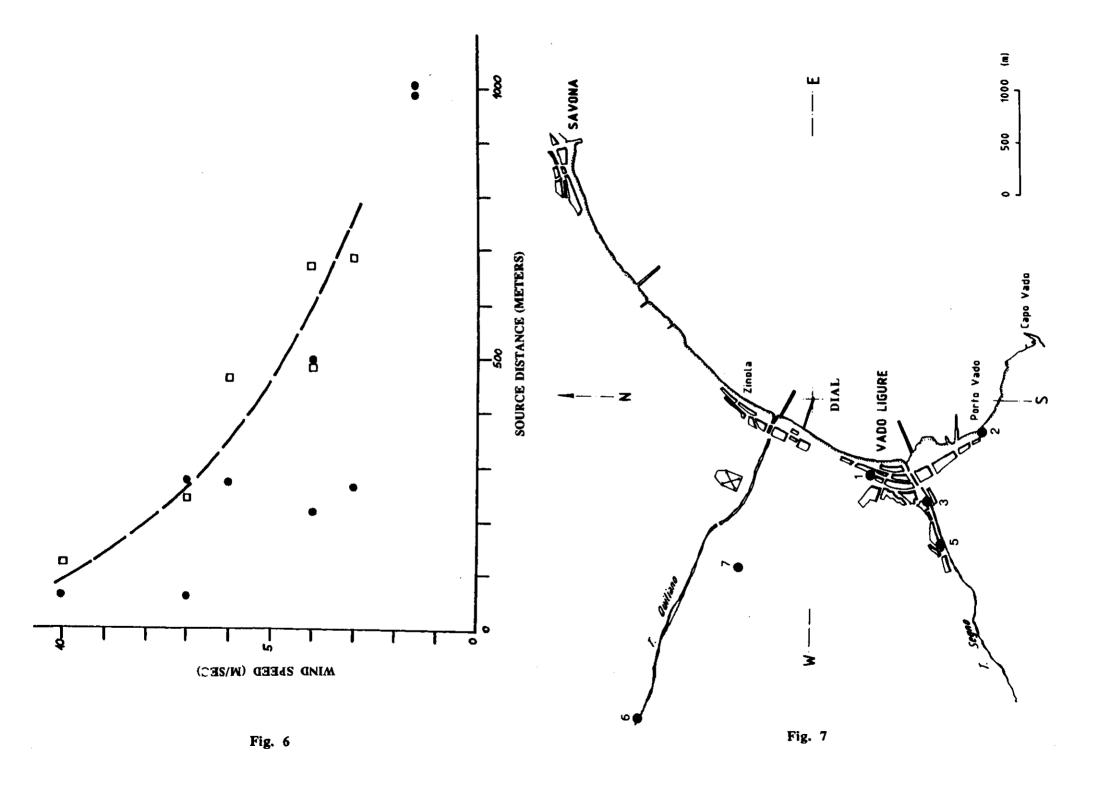


Fig. 5.a

Fig. 5.b



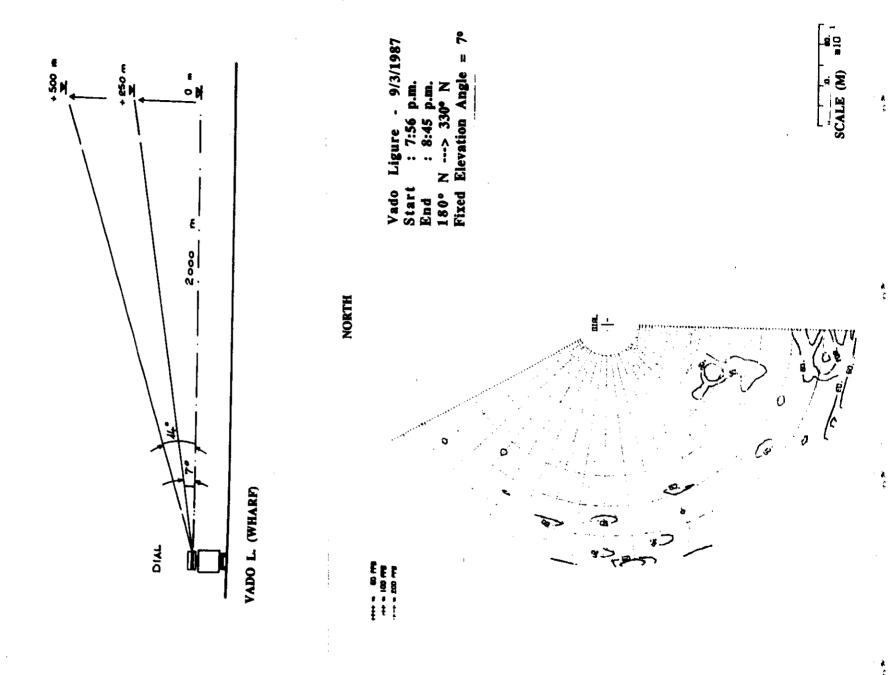


Fig. 8

Fig. 9.a

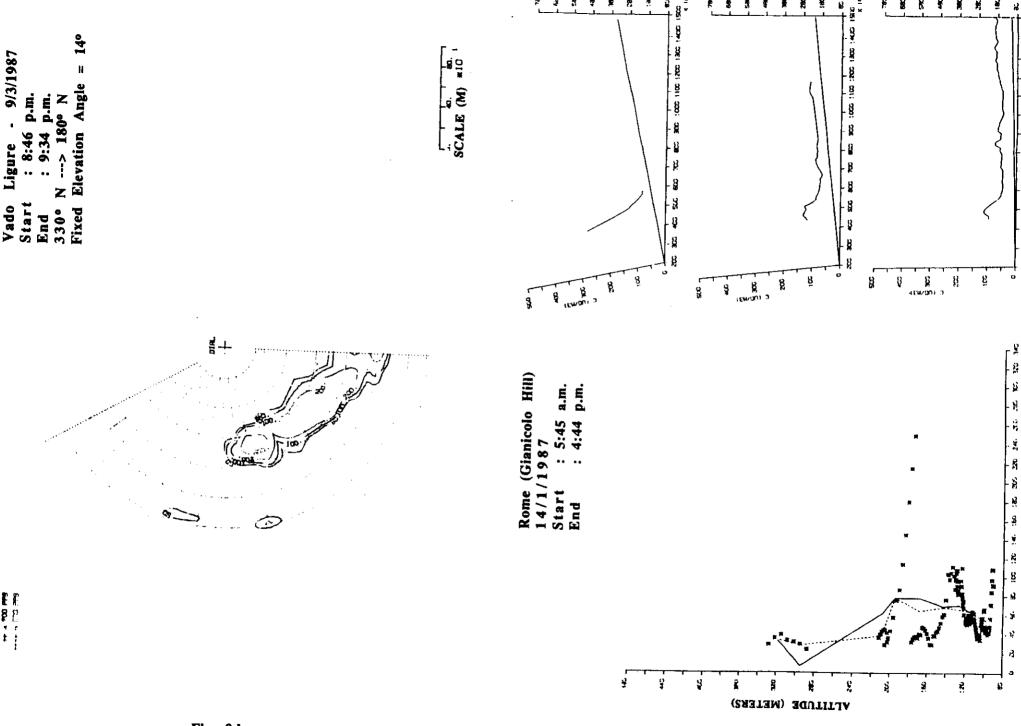


Fig. 9.b

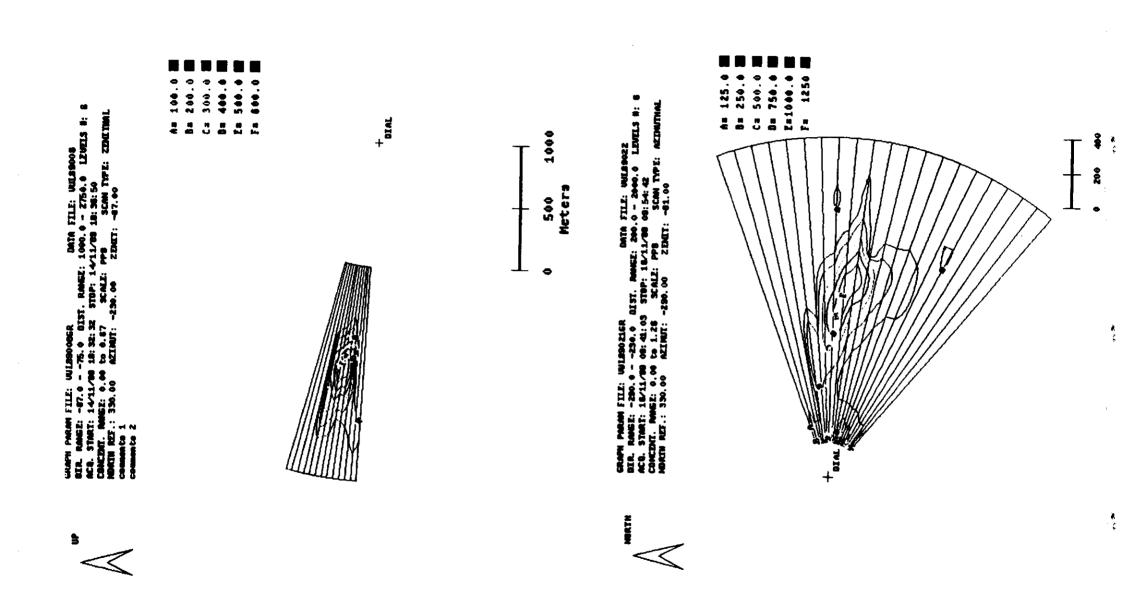


Fig. 11.b

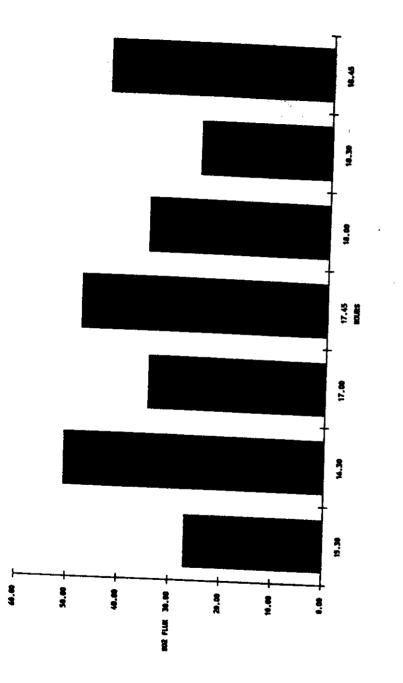


Fig. 12

