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ABSTRACT

The application of a mobile differential absorption lidar (DIAL) system in monitoring the atmospheric distribution of atomic mercury is discussed. The DIAL technique using the 253.65 nm Hg resonance line has been employed in studies of industrial emissions as well as geophysical manifestations. Hg concentrations down to the background value 2 ng/m^3 can be measured.

1. INTRODUCTION

There is a considerable interest to obtain a better understanding of the magnitude and the distribution of different natural and anthropogenic sources of atmospheric mercury, and to study the fluxes between different systems in the environment.[1,2] Mercury is released e.g. from chlorine-alkali industries, refuse incineration plants coal-fired power plants and crematoria. Mineralizations, geothermal reservoirs and volcanism are also known to be associated with elevated mercury concentrations. It has been suggested that geothermal energy and ore deposits might be located focusing on associated atmospheric mercury anomalies.[3,4] Mercury is present in the atmosphere mainly in its atomic form and can thus be detected with the differential absorption lidar (DIAL) techniques using the mercury resonance line at 253.65 nm. During the last years the mobile DIAL system has been used in several measurements of atmospheric mercury in different areas. A Nd:YAG-pumped dye laser with subsequent frequency doubling is used to reach the resonance line with enough pulse energy to enable range resolved measurements up till a range of about 1 km.

2. SYSTEM DESCRIPTION

The mobile DIAL is a flexible system which is constructed for measurements of various species.[5,6] An overview of the system is given in Fig. 1. The laser, telescope and all electronics are contained in a Volvo F610 truck with a specially designed cargo compartment of size 6.0*2.3*2.1 m^3. The truck is equipped with four sturdy supporting legs that can be hydraulically lowered to achieve high stability during measurements requiring high directional accuracy in the optical system. The important characteristics of the DIAL system are summarised in Table I. The laser transmitter is a Nd:YAG pumped dye laser with frequency doubling in beta barium borate (BBO) crystal. The dye laser is equipped with a dual wavelength option, enabling the laser to alternately be fired at two different pre-set wavelengths. The dye laser wavelength scale is calibrated using the absorption in a cell filled with Hg vapour, transluminated with a small part of the laser beam.

The outgoing laser beam is directed coaxially with a vertically mounted telescope and transmitted into the atmosphere via a large flat mirror in a retractable transmitting/receiving dome on the roof. A quartz window seals off the dome. Stepping motors are used to turn the dome and to tilt the mirror. Thus it is possible to steer the measurement direction 360 degrees horizontally and within an angle of 45 degrees vertically. In order to protect the mirror coating, the exiting laser beam is expanded with a 6* beam expander. Servo motors and micrometer screws offer remote control of the final turning prism, through which the overlap between the laser transmission lobe and the telescope field of view is controlled. Two video cameras are used to control and supervise the measurement direction. A mechanical chopper can block the beam if desired. The computer-controlled chopper is automatically used during a lidar measurement to obtain the signal due to background light and preamplifier offset.

An adjustable field stop is placed in the focus of the receiving Newtonian telescope, by means of which the field of view can be varied from 2 to 5 mrad. After passing through an interference filter the light is detected by a photo multiplier tube. Due to the fact that the beam is emitted coaxially with the telescope, the near field back scattered light is very strong. To prevent detector overload and to reduce the dynamic range of the signal, the gain of the PMT is modulated. This is performed by changing the voltage difference in the dynode chain. The gain is very low in the beginning and does not

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reach its full value until after 2-10 µs (the rise time is variable).

The signal from the PMT is preamplified and captured with a 8-bit 100-MHz transient digitizer. Normally 2000 channels, each 10ns wide, are recorded, corresponding to a lidar measuring range of 3 km. After the recording, the data are transferred via a GPIB interface to the computer where they are added into a 32-bit data array at a repetition rate of 20Hz. A DIAL measurement cycle consists of 8 shots on each wavelength, fired alternately, and finally two shots with the chopper closed. Several cycles are then averaged and stored on disk after background subtraction. During a measurement normally 100 to 200 cycles are averaged, corresponding to a measurement time of 1.5 to 3 minutes. When vertical or horizontal scanning were performed the measurement time for each direction can be divided in several repetitions.

While system control and on-line data handling are controlled by the system computer, data processing and evaluation can be performed on a separate computer. A typical evaluation consists of three phases. First a Gaussian smoothing function of a few channels width is applied on the raw lidar signals. Then the ratio of the lidar signal at the different wavelengths is calculated. A running average function of adjustable width is finally used to evaluate the concentration as a function of distance. Data from horizontal or vertical scans are presented as a 2D-plot with the concentration value indicated with a grey scale. The data from a vertical scan can also be transferred to a single vertical or horizontal profile with a vertical/horizontal projection.

3. MEASUREMENTS

Many field campaigns have been performed with the mobile system to measure atmospheric atomic mercury from industrial and natural sources.[7-9] Fig. 2 shows a vertical scan through the plumes from a chlorine-alkali plant. The two plumes from the two cell rooms are clearly separated and the flux from the two sourses can be individualy evaluated. The flux is evaluated by multiplying the integrated concentration over the plume cross section area with the wind speed normal to the scan. The total flux was in this example found to be 40 g/h of atomic Hg vapour. In Fig. 3 the distribution of Hg near the cooling towers of a geothermal power plant (30 MW) is shown, Hg concentrations are given in ng/m^3 .[8] The figure is made from horizontal and vertical scans as indicated by the dots. It is interesting to see that the main part of the atomic mercury do not emerge from the top of the cooling tower but rather from the lower part. Fig. 4 shows the vertical scan of an other geothermal power plant (20 MW).[8] The total flux from this plant was evaluated to 20 g/h of atomic mercury. Concentrations down to the background value of about 2 ng/m^3 can be measured. In Fig. 5 the vertical concentration profile over a roasted cinnaber (HgS) deposit at an abandoned mercury mine.[9]

4. CONCLUSION

As demonstrated by the examples the differential absorption lidar method is a powerful technique for mapping of important atmospheric pollutants. The atomic mercury measurements were supported with gold trap point monitors and the agreements between the two methods were very good.[10] For the future we plan to extent our investigations of atmospheric atomic mercury to seismic and vulcanological processes, which are also known to produce mercury anomalies.

5. ACKNOWLEDGEMENTS

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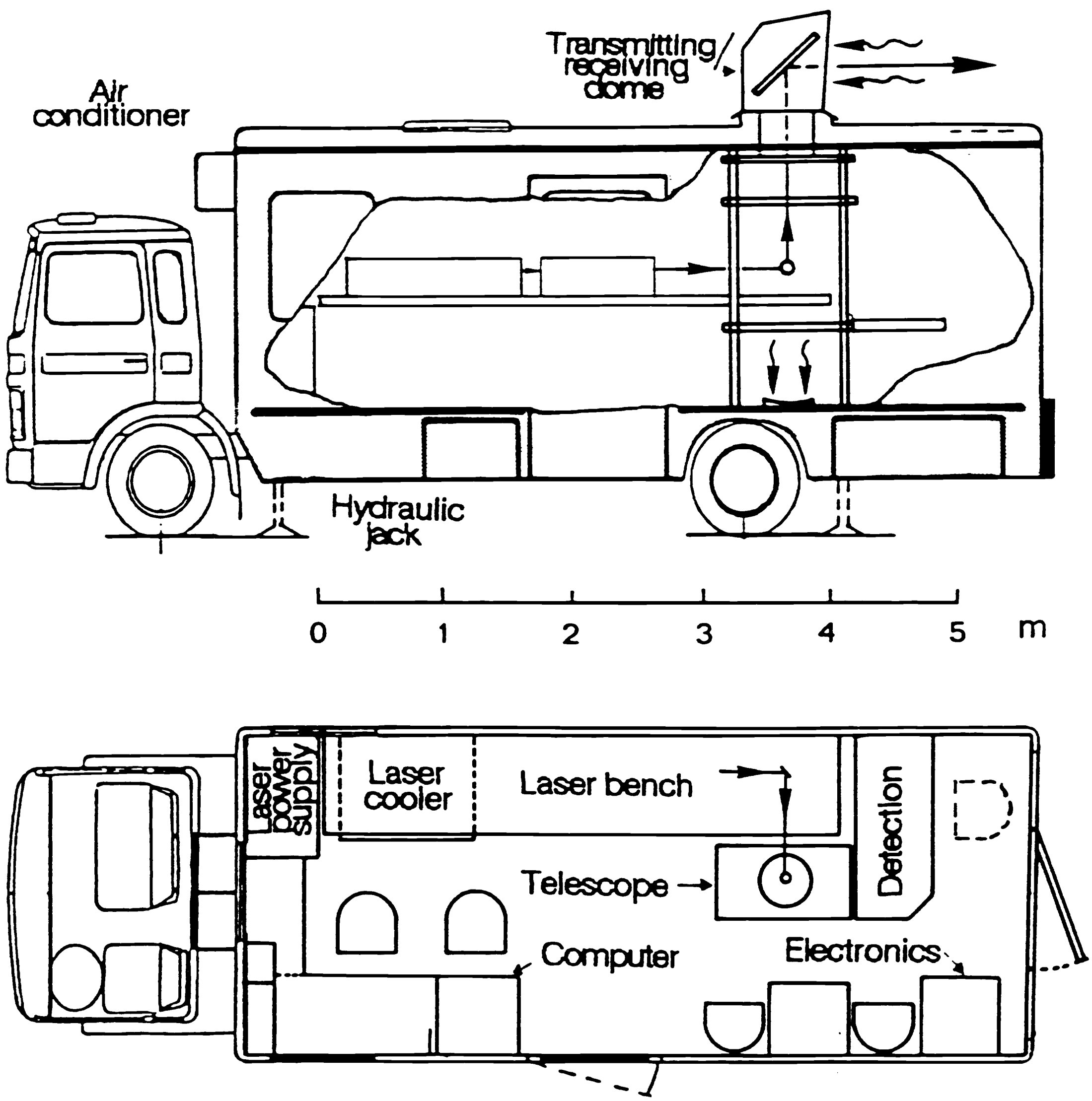

Figure 1. Overview of the mobile DIAL system.

Table I Data for the mobile DIAL system

Emitter		Reciver	
Laser	Continuum, YG682-20, Nd:YAG rep. rate 20 Hz, 6-9 ns pulse length 1200 mJ 1064 nm	Telescope	Newtonian telescope, 40 cm diameter, f/2.5 computer controlled steering mirror
		Photo detector	Thorn-EMI 9816QA, S20 cathode gain 10^7 , ramped 2-10 μs
Dye laser	Continuum, TDL60 frequency doubling 253.65 nm, 5 mJ, 0.5 mrad dual wavelength, alternate switching	Digitizer	LeCroy units 6102, 2*TR 8818 2*NM8103A, 8013. 8-bit, 100-MHz
		Computers	2*IBM compatible PC, 386

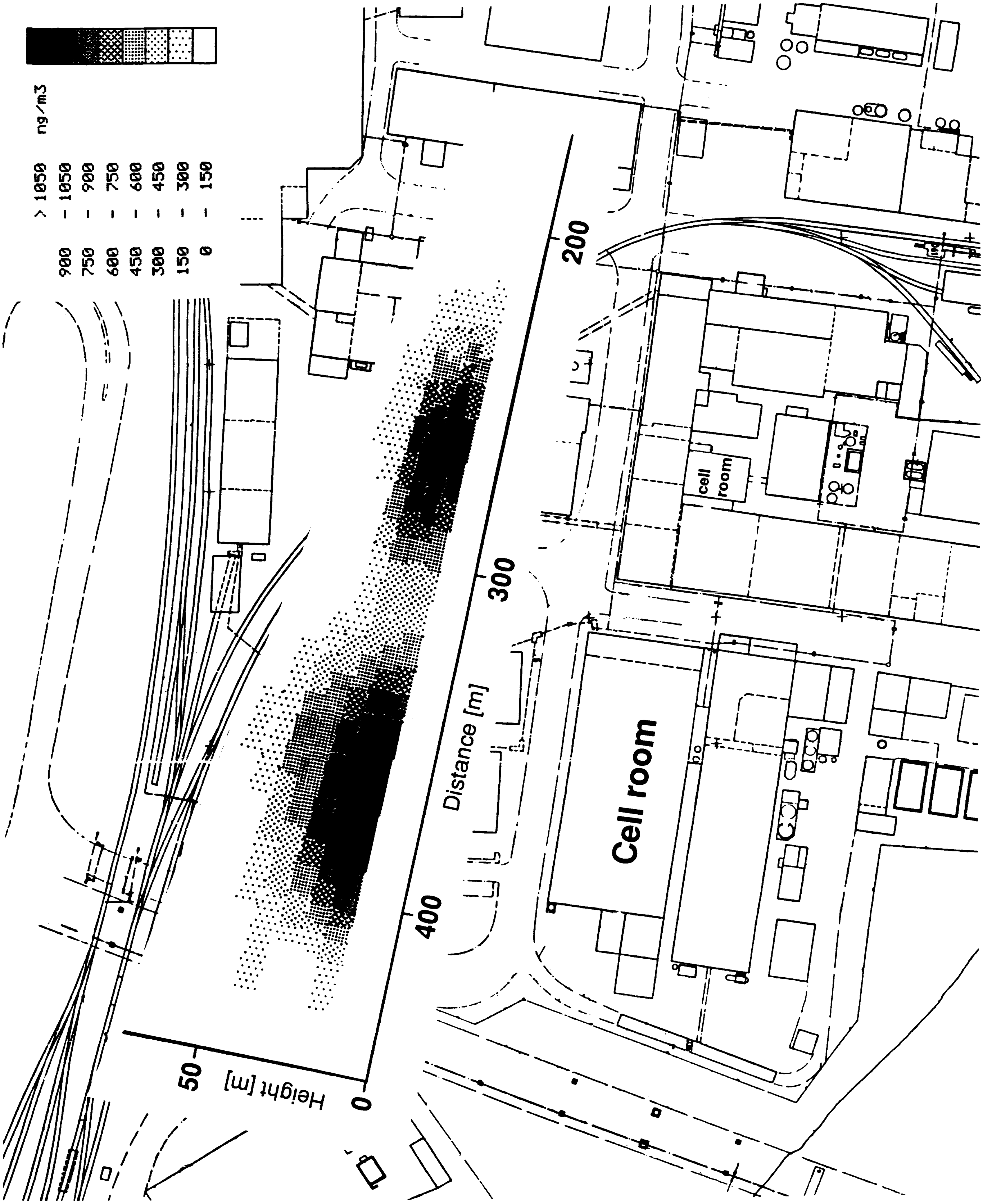

Figure 2. Vertical scan of the Hg plumes from a chlorine-alkali plant.

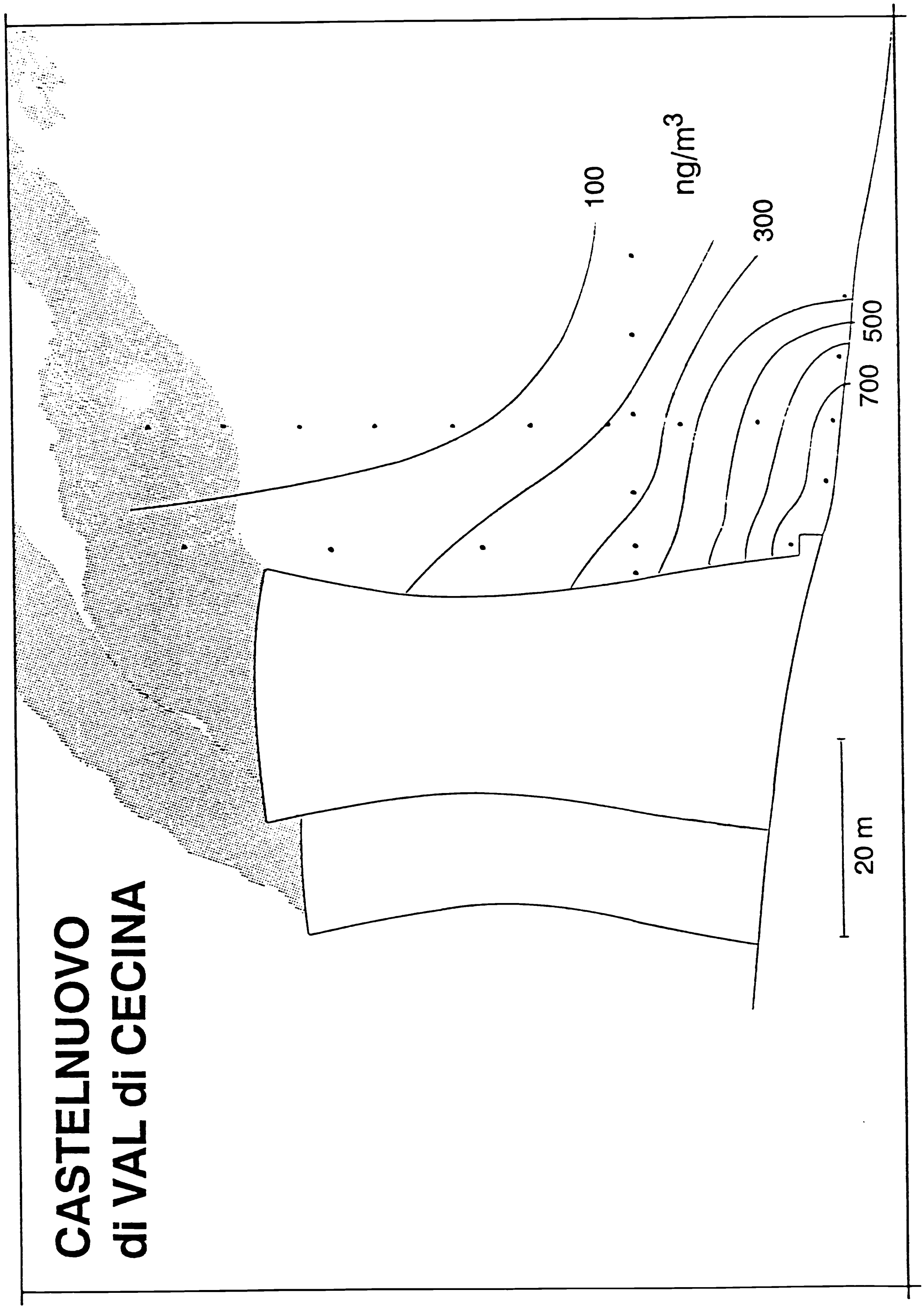

Figure 3. Hg mapping close to the cooling towers at a geothermal power plant, Hg concentrations in ng/m^3.

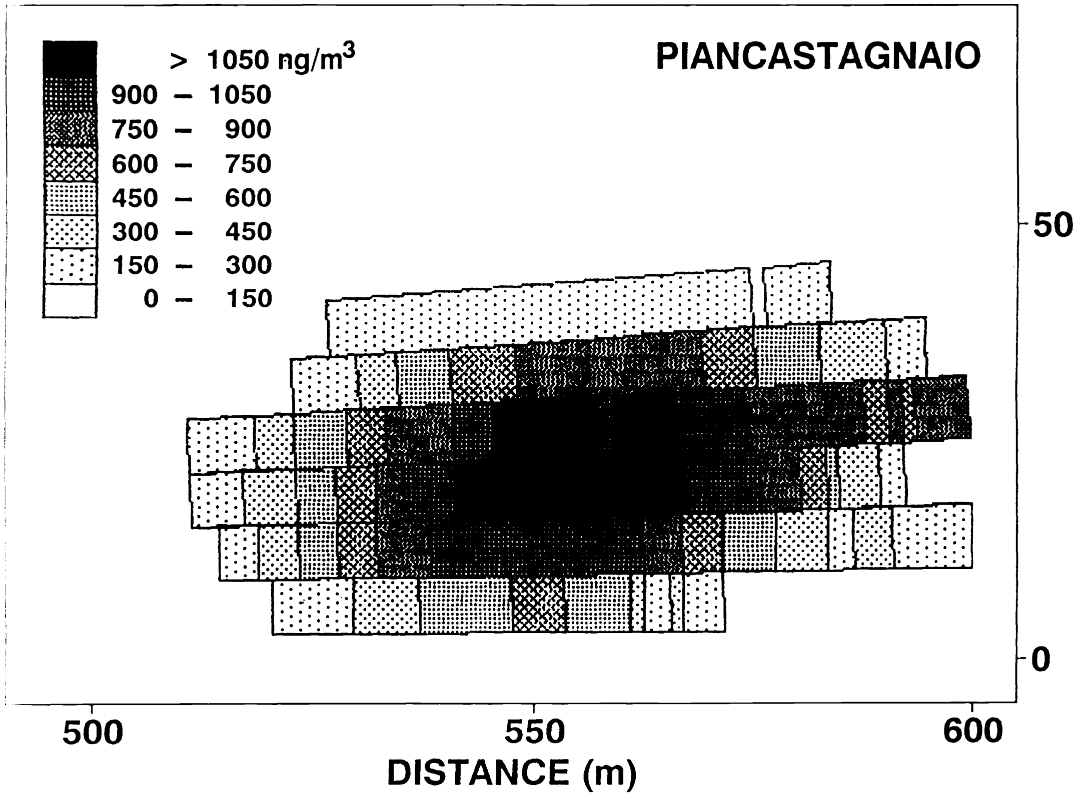

Figure 4. Vertical scan of the Hg plume downwind a geothermal power plant.

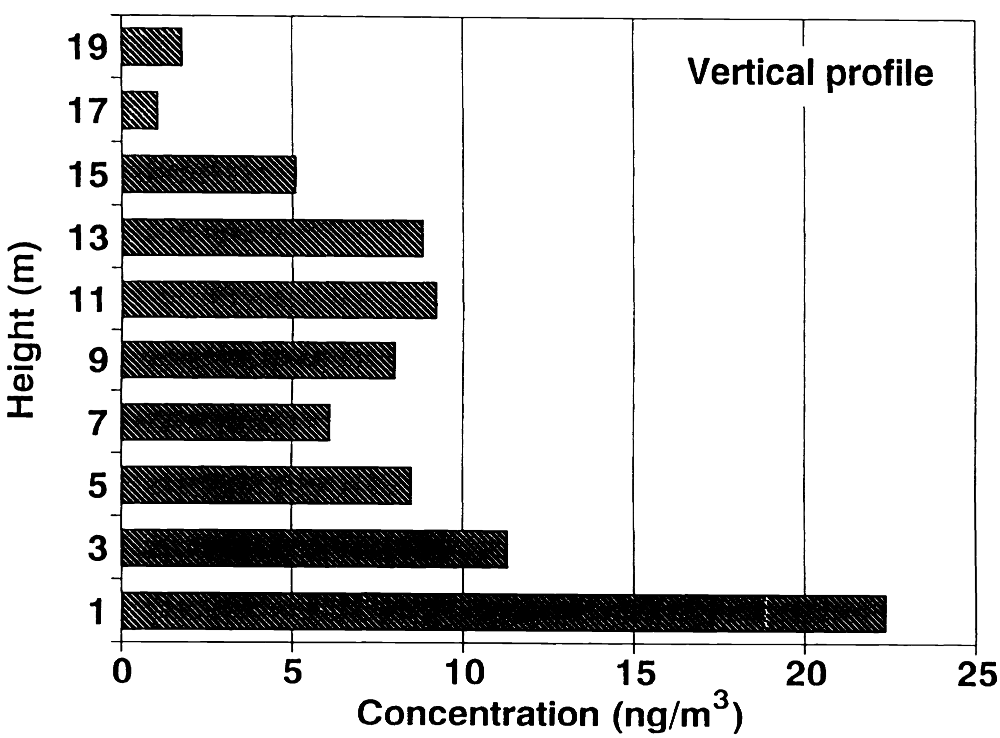

Figure 5. Vertical profile of Hg over a roasted cinnaber deposit.