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**A SYSTEM FOR MEASURING FLUXES OF  
TRACE GASES TO AND FROM SOIL AND  
VEGETATION WITH A CHAMBER  
TECHNIQUE**

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A SYSTEM FOR MEASURING FLUXES OF TRACE GASES TO AND FROM  
SOIL AND VEGETATION WITH A CHAMBER TECHNIQUE

by

Christer Johansson, Andreas Richter, Leif Bäcklin and  
Lennart Granat

ABSTRACT

A mobile system for measurements of gaseous fluxes of  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and water vapour between the atmosphere and the vegetation/soil system using a chamber technique is described.

Equipment, instruments and instrumental sensitivities are listed. A gas-handling system including calibration procedures and information on the chamber construction is also presented.

With this system it is possible to make measurements using a wide range of concentrations including subambient levels. The environment in the chamber can be kept very close to ambient conditions. The mechanisms regulating the flux can be studied. A limitation of this system is the problem of extrapolating the fluxes obtained to larger areas.

CONTENTS	PAGE
INTRODUCTION	3
IMPLEMENTATION	3
EQUIPMENT AND MEASURING SYSTEM	6
Instruments and gas-handling system	6
Flux measurement chamber	7
Calibration and permeation tube system	8
CONCLUDING REMARKS	9
REFERENCES	10

## INTRODUCTION

In recent years the exchange of gaseous compounds between the atmosphere and different types of surfaces has received increasing interest, not the least in connection with studies of large-scale pollution problems and biogeochemical cycles. Two important fluxes in this connection are dry deposition and biogenic emission. There have been several efforts to measure and estimate dry deposition (e.g. Garland, 1978; Fowler, 1978) as well as biogenic emissions of reduced sulphur compounds (Aneja, 1981) and  $\text{NO}_x$  (Galbally and Roy, 1978). These measurements are, however, limited to a few geographical sites. At this institute, we have also investigated problems concerning flux measurements of trace gases and some measurements have been performed (Richter and Granat, 1978; Hällgren et al., 1982; Johansson and Granat, 1982; Johansson et al. 1982; Johansson and Granat, 1983) as well as estimates of dry deposition (Granat, 1982; Söderlund, 1981). Our earlier measurements indicated that there are variations in dry deposition/emission rates due to abiotic and biological variables within the ecosystem. For example, large variations in the flux of trace gases are caused by variations in species, nutrient supply and climate. This must be remembered when selecting suitable measuring techniques and estimating procedures. In this report we describe a mobile system for simultaneous measurements of fluxes of  $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and water vapour using a chamber technique.

## IMPLEMENTATION

Among the different methods available for dry deposition and emission studies (see for instance Hicks et al. (1980) and Sehmel (1980)) we have chosen the chamber technique. It has previously been used in field experiments by among others Linder et al (1980); Milne et al. (1979) and Hällgren et al. (1982).

With this method a transparent chamber is mounted on selected vegetation elements or placed on different ground surfaces. In an open chamber system, controlled concentrations of trace gases at ambient or any predetermined fumigation level are distributed to the chamber with the aid of a gas-handling system. Fluxes to or from the investigated surface are derived from measurements of concentration difference between the inlet and outlet of the chamber using information on the air flow rate

and surface area. Great care has to be exercised in checking uptake in tubings and on the walls of the chamber during different environmental conditions. The chamber technique is not restricted to an open system but can also be used as a closed system when the flux is calculated from changes in concentration with time. The chamber technique offers several advantages.

- It is a direct measurement technique since the deposition/emission rate is directly calculated from the concentration measurements;
- Mechanism studies can be performed since the fluxes of trace gases and different biological processes (e.g. photosynthesis and transpiration) can be carefully examined simultaneously;
- The method offers great possibilities for effect studies (dose/response relationship) since the concentrations of different trace gases, for instance toxic or potentially acidifying substances, can be controlled and adjusted;
- Furthermore, the climate in the chamber can be varied over a wide range of conditions. It is, for example, possible to study the processes occurring on wetted surfaces by removing the chamber during a precipitation event or by artificially introducing rainwater of different chemical composition. Alternatively, the chamber can be removed from the branch in order to allow normal rainfall to penetrate the foliar surface;
- The chamber technique can also be used in the laboratory.

However, for the deposition estimate with the chamber technique to be representative for the forest canopy, it has to be assumed that the aerodynamic and boundary layer resistance can be neglected compared to the surface resistance. Inside the chamber the boundary layer resistance can be considered negligible due to the use of a propeller.

Concerning the surface resistance for  $\text{SO}_2$ , the stomatal resistance is considered to be the dominant factor regulating deposition and emission processes, but both an internal resistance (Hällgren et al., 1982) and a cuticular resistance (Fowler, 1978) have been shown to exist sometimes.

The stomatal resistance for water vapour can be calculated from dew-point measurements at the chamber inlet and outlet and by measuring the needle temperature as described by Lohammar et al. (1979).

By combining these resistances according to the electric resistance analogy, an overall mass transfer resistance  $R_g$  can be defined as

$$R_g^{-1} = \frac{1}{r_a + r_b + r_s}$$

where  $\frac{1}{R_g}$  is often called deposition velocity,  $V_g$ , and where  $r_a$ ,  $r_b$  and  $r_s$  are the atmospheric, boundary layer and surface resistances. Assuming the flux to be proportional to the concentration in the chamber, we can write the following equation

$$F = V_n \cdot C$$

where  $F$  = flux of the gas to the surface,  $V_n$  = deposition to the surface area in the chamber and  $C$  = trace gas concentration in the chamber.

The deposition calculated above can, however, not be transformed to an overall dry deposition velocity,  $V_g$ , for the whole ecosystem in question without detailed information on the temporal and spatial variability of the dry deposition velocity. A first approximation often used is to assume that  $V_g$  is proportional to  $V_n$

$$V_g = (\text{LAI}) \cdot V_n$$

LAI = leaf area index (projected surface area/ground area).

Interesting information can be achieved by comparing the calculated surface resistance from the flux measurements of a compound and the stomatal resistance for the same compound calculated from water vapour measurements. The stomatal resistance also provides information on the status of the surface and can for example be used as an indication of water stress (Bengtson, 1979; Larsson, 1979).

More theoretical aspects of the chamber technique are given by Rogers et al. (1977) and Unsworth (1982).

As with other methods, there is still a problem of verifying the measure-

ments but comparison with eddy flux correlation measurements, the gradient method, throughfall data or comparison with the catchment area technique might serve as helpful tools for verification.

#### EQUIPMENT AND MEASURING SYSTEM

The measuring equipment is installed in a mobile van (Polar 420T, Sweden) easily pulled by a car (see Fig. 1).

The instruments are placed in racks in the middle of the van making it possible to reach both front and rear sides of them. The pumps are placed in a box mounted on the outside of the van in order to minimize vibrations and noise. The inside temperature of the van is considered as an important environmental factor for the instruments described below and in order to keep the temperature constant, air conditioning has been installed.

#### Instruments and gas-handling system

A general outline of the gas-handling system is given in Fig. 2 and the instruments are listed in Table 1. Ambient air is first pumped (GAST, DOA101, Gast Manufacturing Corporation, USA) through filters (Whatman 40 mm, impregnated with approximately 0.5 ml 0.5 M KOH) mounted in filter holders ("SWINEX", polypropylene) which remove SO<sub>2</sub>. Since the removal efficiency of SO<sub>2</sub> depends on the relative humidity of the air, the filters are placed on the outside of the van. To maintain removal capacity, the filters are replaced every week.

The air coming into the van at a selected flow rate (often 10 l min<sup>-1</sup>) is divided into three separate airflows. The first is controlled by a massflow meter (Brooks, Thermal flow controller 5811, USA) and leads to a thermostated ( $\pm 0.01^{\circ}\text{C}$ ) water bath (Lauda Kryostat, FDR) containing the permeation system described below. The second airflow is periodically passed directly to the instruments giving a zero signal from the analysers. The third airflow is fed to the flux measurement chamber chosen in the experiment. Any concentration level can be achieved by mixing line one and line three. A small portion of the air administered to the chamber is periodically passed to the instruments to check the concentration before the chamber. The air is then pumped from the chamber using a special pump

with teflon membrane (MB 41E Metal Bellows Corporation, USA) and through a pressure equilibrating teflon vessel to the continuous analysers. The airstreams are all connected by one-way teflon solenoid valves. The length of the measuring periods is controlled by a micro computer (ABC-80) which is also used for recording and data logging. The micro computer processes and stores data on magnetic discs and prints out the preliminary results. For instantaneous data checking strip chart recorders are used. All tubings are made of FEP teflon (4.2 mm HABIA, Sweden) and they are all connected with polypropylene fittings. The tubings leading to and from the chamber are heated and insulated in order to prevent condensation of water vapour on the walls.

#### Flux measurement chamber

Both chambers described below are constructed at the Institute. The chamber used for branches, shoots and small plants is shown in Fig. 3. The chamber material is a thin (0.025 mm) transparent (90% transmittance between 100 and 300 nm) FEP teflon film. The volume of the chambers varies between 5 and 10  $\ell$ . The chamber is mounted around the shoot which is then sealed by tightening of the teflon film around the stem of the branch. In order to eliminate the boundary layer resistance and to promote rapid mixing of the air in the chamber, a teflon propeller (5 cm diameter) driven by an external motor is used. Chamber air temperature is measured with a temperature sensor placed in the outlet of the chamber and shielded against radiation and well ventilated.

A larger plastic bag placed around the chamber makes it possible to control the temperature within the chamber. For example, if the dew point measurement indicate that dew formation might occur on the chamber walls, heated air can be blown into the space between the teflon chamber and the plastic bag. This airstream can be supported automatically by a separate fan and the air is heated by a resistive coil placed in the airstream. The light intensity sensor is placed close to the flux measurement chamber.

The chamber constructed for ground surface measurements is shown in Fig. 1. It consists of a transparent teflon film mounted on the inside of an aluminium frame. The volume is approximately 100  $\ell$ . The air inside



the chamber is mixed with a stainless steel paddle (see Johansson and Granat, 1983).

#### Calibration and permeation tube system

The SO<sub>2</sub> and NO<sub>x</sub> analysers are calibrated by the use of permeation tubes. The permeation tubes (steel body with a teflon membrane) were constructed at the laboratory and filled with liquid SO<sub>2</sub> and NO<sub>2</sub>, respectively. At 25°C the permeation rate can be varied from 1.0 to 10 µg h<sup>-1</sup> depending on the thickness and diameter of the membrane used. Calibration curves in the range 0.5 to 50 ppb can be obtained by regulating the airflow rate over the permeation tubes. The NO<sub>x</sub> analyser is also calibrated by using NO in N<sub>2</sub> mixtures (1 to 50 ppbv NO in N<sub>2</sub>) from gas cylinders. Since the instruments exhibit zero and span drift they are span-checked every hour. The SO<sub>2</sub> analyser shows interference from changes in atmospheric pressure, CO<sub>2</sub> concentration and water vapour content (Tanner et al., 1980). The interference from pressure changes has been eliminated in our gas-handling system since the measurements are always performed at atmospheric pressure. Earlier measurements indicate a negligible CO<sub>2</sub> difference between inlet and outlet of the chamber regarding the influence on the SO<sub>2</sub> measurements (Eaton, 1978). A correction curve for the interference from water vapour has been established at the laboratory.

The O<sub>3</sub> analyser is calibrated with the aid of an external O<sub>3</sub> generator which is wet-chemically calibrated at the laboratory. The calibration of the dew point analyser is checked at the laboratory. Sensor cleaning is regularly required to maintain proper operation. The temperature measuring device is constructed at the laboratory and its linearity and absolute reading have been checked (ice/water mixture, waterbath and boiling water). Soap bubble meters and a wet test meter are periodically used for the calibration of both rotameters and massflow meters. For the light intensity sensor we use the factory calibration. The total projected leaf area is measured after each set of measurements with a leaf area meter (Lambda Li 3000, Lambda Inst., USA).

The mobile system described above has been used for dry deposition and emission measurements at different field sites and during different periods of the year. Results have been reported in Johansson and Granat (1982 and 1983) and Johansson et al. (1982).

#### CONCLUDING REMARKS

Many of the fumigation experiments performed earlier with the chamber technique have suffered from unrealistic high concentrations of the pollutants studied. With the system described in this report, it is possible to achieve a wide range of concentrations including subambient levels.

Although the chamber technique to some extent offers an artificial environment for the enclosed shoot or ground surface, environmental variables such as temperature, relative humidity and light intensity can be measured and kept very close to ambient conditions. We therefore believe that the measuring equipment can give reasonable estimates of the flux of trace gases to or from the investigated surface within the chamber. Interesting studies on the mechanisms regulating uptake or emission can also be performed. The major problem with the method is the question of representativity of the measurements and the extrapolation of the fluxes obtained to larger areas.

Systematic comparisons between flux chamber measurements and measurements obtained with other methods, e.g. eddy correlation technique, are highly desirable.

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Table 1. Measuring equipment.

Measured parameter	Equipment	Sensitivity	Working principle
SO <sub>2</sub> (total sulphur)	Meloy SA285, modified Meloy Lab. Inc. (USA)	0.3 ppb	Chemiluminescence Flame photometer (reaction with hydrogen)
NO and NO <sub>x</sub> (includes NO, NO <sub>2</sub> , PAN and several nitrates i.e. HNO <sub>3</sub> )	Thermo-Elektron Co. Model 14D, modified (USA)	0.1 ppb	Chemiluminescence (reaction with O <sub>3</sub> )
O <sub>3</sub>	Constructed at the Dept of Meteorology, University of Stockholm, Sweden	2 ppb	Chemiluminescence (reaction with ethylene)
Water vapour	Dew-point hygrometer 880, EE86 Int. Inc. (USA)	0.3°C	Cooled dew point mirror
Temperature	AP590, Analog Device, (USA)	0.05°C	Semiconductor
Light intensity	Lambda, Li-130S Lambda Inc. (USA)		Silicon photodiode

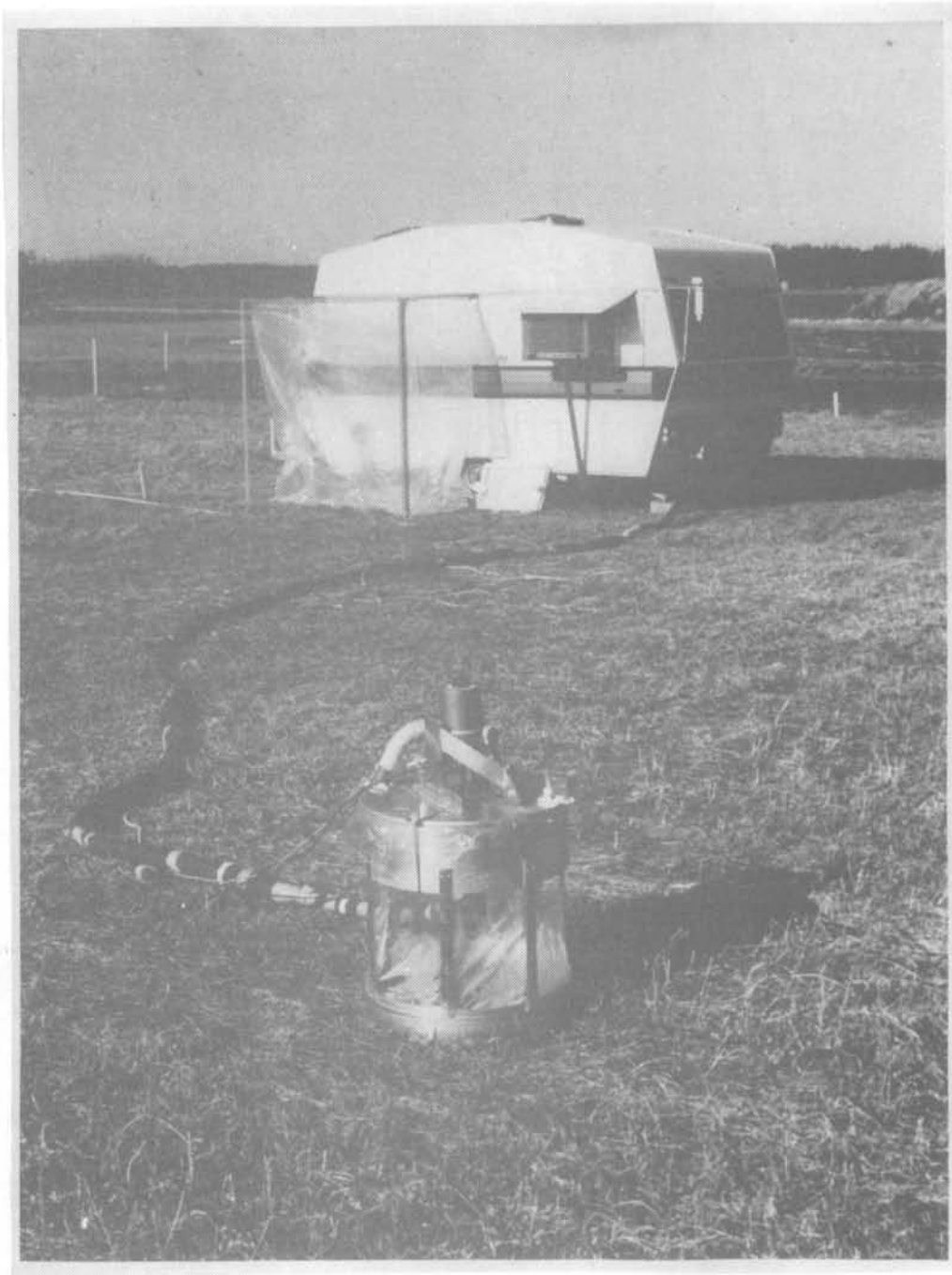


Figure 1. The mobile van and the chamber for ground measurements

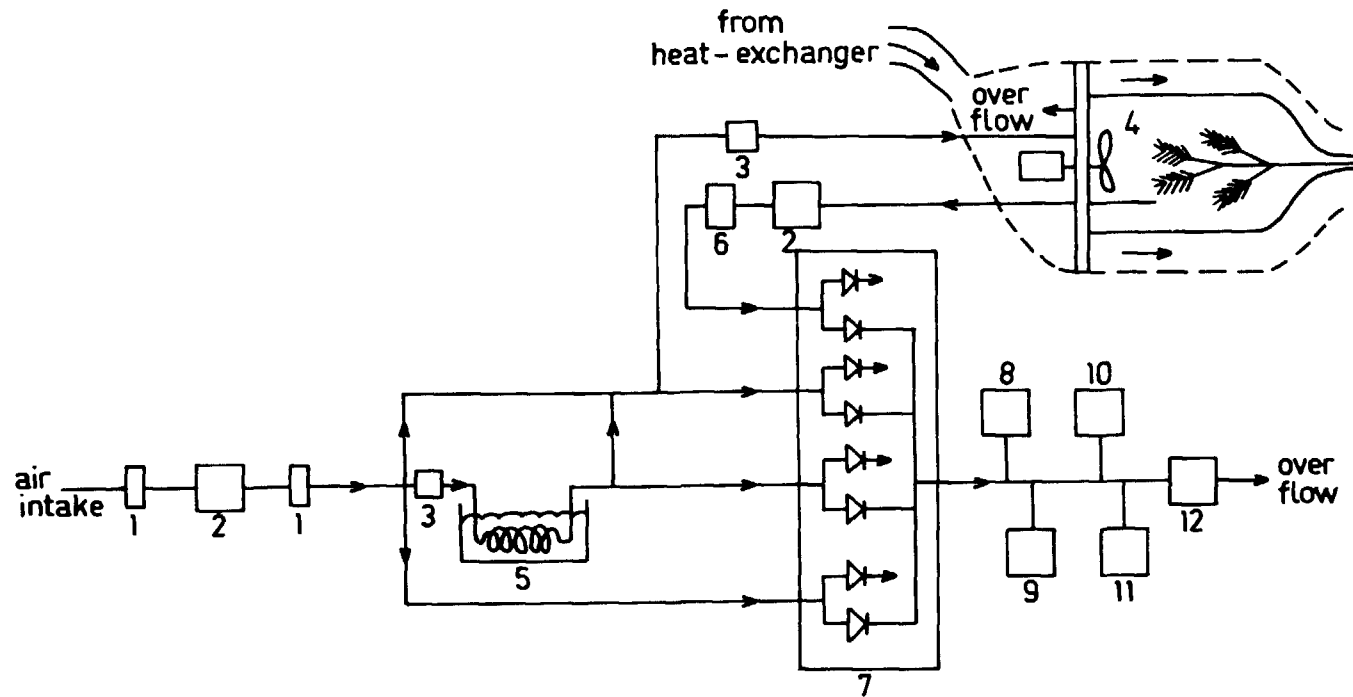


Figure 2. Schematic description of gas-handling and measuring system.

- |  |                                     |
|--|-------------------------------------|
| 1. Filters                             | 5. Water bath with permeation tubes |
| 2. Pump                                | 6. Pressure equilibrating vessel    |
| 3. Mass flowmeter                      | 7. Solenoid valve system            |
| 4. Flux measurement chamber with shoot | 8-12. Continuous analysers          |

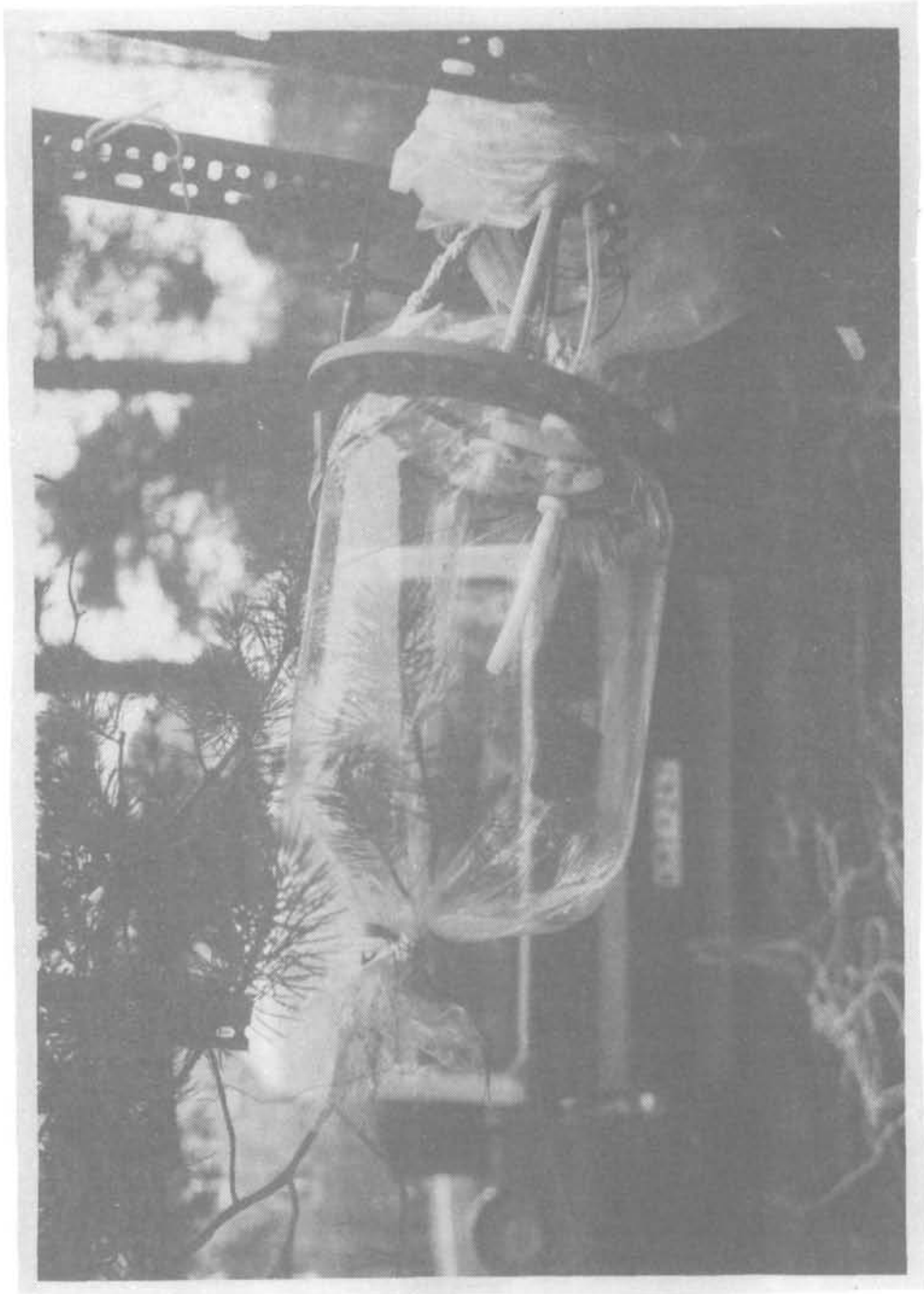


Figure 3. The chamber for measurements of trace gas fluxes to or from vegetative elements.



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