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David Pinkston

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A Study on Gaseous Ebullitions of
De Gray Reservoir, Arkansas

submitted by David Pinkston in partial fulfillment
of the requirements of the University Honors Program

Honors Director: Dr. Joe Jeffers

May, 1979

Acknowledgements

Thanks go to Dr. Joe Jeffers for his worthy advice, assistance, and financial support. Great thanks also go to Dr. Joe Nix, whose financial support and counsel enabled this project to be conducted. All the members of NART helped often and were essential to the continuation of this project. Thanks also go to Ralph Roseberg for use of water column profile data.

This study was undertaken in order to determine the nature of and the factors that influence the gaseous ebullitions of De Gray Reservoir near Arkadelphia, Arkansas during a yearly cycle.

Two major types of gaseous ebullition are described in the literature. The first has to do with the production and ebullition of elemental oxygen from photosynthesis (Porcella et al., 1975). Hutchinson (1957) predicts that the formation of oxygen bubbles in this manner is limited to very shallow, well lighted, warm waters.

The second type of gaseous ebullition involves the anaerobic digestion of organic matter (Reeburgh, 1969) (Howard et al., 1971). Anaerobic conditions exist in the hypolimnion during summer stratification and usually in sediments, even when they are covered by well aerated waters (Porcella et al., 1975). The silt-absorbed organic matter undergoes anaerobic breakdown to form fatty acids, which are then converted mainly to methane (Kuznetsov, 1968). Also produced are ethylene, ethane, carbon dioxide, nitrous dioxide, propane, and higher molecular weight hydrocarbons (Smith et al., 1969). Although ethylene is produced in quantities second only to methane, it is quite soluble in water, and it is apparantly a better substrate for bacteria than is methane (Porcella et al., 1975). Hydrogen sulfide is also produced, but it is often precipitated as iron sulfide in anaerobic waters (Reeburgh, 1969).

The methanogenes are bacteria responsible for most of the methane production (Buchanan, Gibbons, 1974). Gram

negative methane producing bacilli have been isolated from methane evolving sediments in Lake Erie (Howard et al., 1971)

The methane produced forms into bubbles only when the partial pressure of the gas exceeds the in situ pressure (Klots quoted by Reeburgh, 1969). For a lake at sea level at a depth of 10m and a temperature of 7°C the critical concentration for bubble formation is between 26 and 38 ppm methane, while at a depth of 20m and a temperature of 4°C the critical concentration lies between 52 and 76 ppm methane. Thus the production of methane bubbles up to depths of 10m is reasonable but is unlikely to take place at depths approaching 20m except in very polluted waters where anaerobic digestion activity is higher than usual. Rossolimo reported maximum ebullition from depths between 9m and 10m at Lake Beloje near Kossino, Italy in the summer of 1935 (Hutchinson, 1957). A great deal of methane then simply diffuses from the anaerobic zone. It was estimated that the range of methane lost by diffusion in small, hypereutrophic Lake Wintergreen between May 23rd and August 21st of 1972 was one to two times greater than that lost by ebullition (Strayer et al., 1978). Most of this diffusing methane is oxidized to carbon dioxide by bacteria at the thermocline or during fall overturn (Rudd and Hamilton, 1978) (Kuznetsov, 1968). Methane oxidizing bacteria have been isolated from all parts of the aerated water column, including the sediment surface, in areas of methane ebullition (Howard et al., 1971). Thus methane plays an important part in the lake carbon cycle (Rudd et al., 1974).

It is estimated that the loss of methane from Lake Wintergreen amounts to 11.5 percent of the total organic carbon produced (net productivity estimates including net primary productivity, dark fixation, and excretion) from May to October, 1972 (Strayer et al., 1978).

Studies on gaseous ebullition are not all recent. The data of Rossolimo collected in May of 1935 at Lake Beloje has been used to calculate a rate of ebullition of 15.5 mmole methane/ m².day (Strayer et al., 1978). The data of Howard et al. (1971) gives a rate of 110 mmole methane/ m².day in Lake Erie during August and September of 1969. The gas composition was 95 percent methane, 3 percent nitrogen, and 2 percent carbon dioxide. Lake Wintergreen ebullition from the 30th of May to the 21st of August averaged 21 mmole methane/ m².day while the daily rate maximum measured was 35 mmole methane/ m².day. During this time the composition of the gas was 73 percent or higher in methane with the balance being nitrogen and low quantities of carbon dioxide (Strayer et al., 1978).

Ebullition of nitrogen fits neither of the cases previously considered. Allgeier et al. determined nitrogen ebullition to be about 18 percent of that of methane (volume/ volume) from low temperature fermentation of Lake Mendota mud (Hutchinson, 1957). One process in which nitrogen is evolved involves the stripping of nitrogen from the surrounding water as a methane bubble is formed and/or ascends to the surface. If the surrounding water is saturated with nitrogen, the percentage of nitrogen incorporation into a bubble as

it is formed may be considerable at shallow depths. As the depth (and thus the in situ pressure) increases, the proportion of nitrogen incorporated decreases. (Hutchinson, 1957). As the bubble ascends through water saturated with nitrogen, the bubble may "strip out" a considerable quantity of gas. Rossolimo reports that bubbles formed in Lake Beloje at a depth of 10m contain 74 to 84 percent methane and 5 to 18 percent hydrogen. During the ascent of the bubble all the hydrogen is lost and the methane content decreases to 20 to 24 percent, while there is a great gain in nitrogen and a small gain in oxygen. For this phenomenon to occur, the supernatant water must be well aerated but depleted in methane and hydrogen (Hutchinson, 1957).

Other reports of high nitrogen content in evolved gases such as that of Duggeli, who in 1936 measured 52.9 percent nitrogen in gases from muds of a lake near Lucerne, are explained by nitrogen stripping (Hutchinson, 1957). Selective removal of nitrogen from the sediments during ebullition has been shown by comparing dissolved nitrogen to argon ratios at varying depths in the sediments. Nitrogen is less soluble than argon and is thus stripped preferentially (Reeburgh, 1969).

A second process in which nitrogen is evolved is the microbial production of nitrogen, termed denitrification. Certain organisms use nitrate as a source of oxygen in anaerobic environments and produce nitrous oxide and nitrogen gas among other side products (Porcella et al., 1975). Yet even in sediments where there is unequivocal liberation of nitrogen, the quantities seem to be relatively small when

compared to methane production. In 1932 Allgeier et al. reported finding not more than 0.5ml of nitrogen produced from a liter of Mendota Lake mud in 100 days of anaerobic decomposition at 7°C (Hutchinson, 1957). Yet considerably more nitrate in hypolimnetic waters of Lake Mendota was involved in denitrification (8 to 26 μg nitrogen / 1 day) than in reduction to ammonia or organic nitrogen (1.4 to 13.4 μg nitrogen / 1 day) (Brezonik, 1968).

Several factors influence the ebullition of gases. As previously noted, depth is a crucial factor. Since ebullition is directly related to solubility, and solubility to temperature, temperature would be expected to play a major role. Temperature also plays a major role in determining the activity of most of the organisms involved in gas production (Buchanan, Gibbons, 1974). From the 22nd of February to the 7th of March, gases evolved from a depth of 6m in Lake Wintergreen (average temperature 3°C) amounted to only 4 percent of the average summer ebullition (temperature between 12°C and 16°C) (Strayer et al., 1978).

As expected, the nature of the sediments also influences the composition of the evolved gases and their rate of ebullition. In Lake Erie, rates as high as 110 mmole methane / m^2 , day have been reported over sites well covered with organic sediment while rates at another site only sparsely covered (yet under the same conditions of temperature and depth) were essentially nil (Howard et al., 1971).

METHODS

Gas samples were collected by suspending an inverted funnel over the sediments by an anchored buoy and displacing water from a flask above the funnel (Figure 1). Samplers were set below the thermocline or at 1m to 2m above the sediments where the water column was well aerated. Samplers have been left in place and collected periodically from June, 1978 until the present at one or more of four sites on the upstream portion of De Gray Reservoir (Map 1). These sites were named according to their proximity to previous data collection stations (Nix et al., 1975). One site (near station 14) is in shallow water at the mouth of a small creek. Another site named 14A and two other sites (near station 14 and near station 12) are over the river channel. Sampling periods varied from two weeks during the greatest ebullition activity to almost four months during the least. Dissolved oxygen and temperature profiles of the water column were taken as soon as the gas sampler had been taken out of the water. Samplers were kept in the dark and refrigerated as soon as possible. Sample preservation appears good. A sample taken on the 15th of September was run on the 22nd of September and on the 16th of October with essentially the same results. Recently, filling the collection flask with distilled water before setting it out has cut down on one possible source of error: algal and bacterial growth within the flask.

The composition of the gas sample was determined by gas chromatography. The chromatograph was equipped with

parallel silica gel (39 inch) and molecular sieve 5Å (4 foot) columns and a 2ml gas sample loop. The inlet and column temperatures were maintained at 59°C to 61°C. The carrier gas was helium with a flow rate of 30ml per minute. Detection was by thermal conductivity. A sample injection system involving displacement of the gases by water was devised and later modified to minimize contamination of the sample by air.

Known volumes of each gas were injected for calibration and the area under the chart peak was assumed to be directly proportional to the volume of gas injected. The total volume of each sample was then determined and total millimoles of each gas in the sample were calculated from the peak areas. This value was then converted to mmole/m².day. Temperature and dissolved oxygen readings used for the graphs included in this paper were taken from 10cm to 20cm above the sediment surface.

Chromatographs of certain samples taken from the 14th of June to the 15th of August displayed an unknown "shoulder" on the usual methane peak. Ethane, methane (by Grignard - Ault, 1976), and ethylene (by dehydration - Adams, Johnson, 1949) were synthesized in reasonably pure form to determine the nature of the unknown shoulder. Each was injected individually and then all were injected simultaneously. Retention times easily showed the shoulder to be ethane.

An attempt to determine differences between the bubble composition as it is formed and after it ascends is being

carried out by suspending one sampler near the surface and another near the sediment in the channel at station 14.

RESULTS AND DISCUSSION

No evolved gases were detected at site 12. This agrees well with Hutchinson's previously cited prediction that methane ebullition beyond depths of 10m is not to be expected. The solubility of the gases is simply too high within the range of in situ pressures and temperatures present at site 12 to allow ebullition. Minimum depth and maximum temperature measured coincided on the 15th of September as 12m and 15°C. Maximum depth and minimum temperature measured from the 23rd of June to the present were 16m and 5.2°C.

Many events at the other three sites seem to be closely connected due to their proximity to each other and will be treated together. Each gas evolved will be treated individually. Sampling at station 14 in the channel began on the 14th of June and is the only other site where sampling continues at present. Sampling at station 14 at the creek mouth was discontinued on the 13th of October when the water level became too low. Sampling at site 14A (200m south of 14) began only at peak ebullition and continued only until the 3rd of November. Due to the fact that values obtained for evolved gases are only averages over the collection periods, only major yearly trends can be seen. An attempt to correlate storm events with gas ebullition was not successful.

At both sites near station 14 an initial high ebullition of nitrogen (in the channel much greater than any other gas) followed by a leveling off period is seen (graphs 3T and 2T). As the temperature increases, the solubility of the dissolved gases decreases until the critical concentration is reached and bubbles are formed. Indeed a seasonal warming of 10°C could cause dissolved gases to exceed the critical concentration by as much as 30 percent (Reeburgh, 1969). Formation of nitrogen bubbles or simply stripping of nitrogen by methane bubbles as previously described would explain these high values for nitrogen. At the time of this writing, an experiment which may distinguish between the two is in progress. This experiment may also distinguish between nitrogen produced by denitrification and that produced by stripping.

The ebullition of methane seems mainly to be temperature dependent (graphs 1T, 2T, 3T). As temperature increases, the initial nitrogen peak falls and methane ebullition increases to a maximum. Yet since in situ pressure and temperature above the sediments is depth dependent, methane ebullition is also depth dependent (graphs 1D, 2D, 3D). Temperature does seem to be the limiting factor. In the creek mouth at 14, temperature and depth both decreased during the first half of October, and methane ebullition drastically decreased (graphs 3T, 3D). Also from the 13th of October, 1978 to the 21st of February, 1979 the depth at the channel site near station 14 remained essentially constant at a shallow 4m while the temperature

above the sediments fell from above 22°C to below 5°C. Methane ebullition fell from 30.5 mmole/m².day to nil during this same time period (graphs 2T, 2D). If this dependence of methane ebullition on temperature holds, a shallow site should reach maximum ebullition as temperature increases before a deeper site. This is indeed the case if one consults graphs 2T and 3T. Due to various sampling problems the period of maximum ebullition is not clearly known for the channel site, yet graph 2T shows this period to be between August 15th and November 3rd (maximum ebullition measured was 30.5 mmole methane/m² day from the 13th of October to the 3rd of November). At the shallower site maximum ebullition is essentially reached by the 2nd of August (94 mmole methane/m².day from the 23rd of July to the 2nd of August with maximum measured ebullition at 98 mmole methane/m².day from the 1st to the 15th of September). Optimum sediment temperature for methane ebullition seems to be at least above 20°C which corresponds well with what is known about methanogenes (Buchanan, Gibbons, 1974).

In observing graphs 1T and 1D one sees that the dependence of methane ebullition on temperature and depth also holds for site 14A. Temperature data from the channel site at 14 (only 200m north of 14A and at the same depth) (graph 2T) is useful in determining that the temperature near the sediment water interface is actually falling during the sampling period in question.

Average methane ebullition over the channel site at

station 14 was 16.6 mmole/m².day from the 6th of July to the 13th of October, 1978. At site 14A this average value was 22.2 mmole methane/m².day from the 1st of September to the 3rd of November. These values are comparable to the previously cited 15.5 mmole methane/m².day obtained from the data of Rossolimq collected in May of 1935 at Lake Beloje. They are also on the same order as the 21 mmole methane/m².day average obtained from the 30th of May to the 21st of August, 1972 at Wintergreen Lake (Strayer et al., 1978). An average value of 72.1 mmole methane/m².day was obtained for the creek mouth site at 14 from the 14th of June to the 15th of September. This is comparable to the value of 110 mmole methane/m².day obtained by Howard et al. (1971) in Lake Erie during August and September. Methane ebullition was negligible at the three sites or samples were not taken during the periods not included in the averages.

Dissolved oxygen in the supernatant waters was seen to have little effect on methane ebullition from the sediments. The dissolved oxygen data is perhaps misleading in that readings for the graphs were taken anywhere from 10cm to 20cm above the sediments (a zone of very rapid dissolved oxygen change). Of course, dissolved oxygen is also greatly dependent on depth and temperature. Graph 2.0 shows methane ebullition at low dissolved oxygen readings and at high dissolved oxygen readings. Graph 3.0 shows dissolved oxygen remaining high while methane ebullition increases and then decreases from June to October.

of ethylene produced in their anaerobic microcosms. The ebullition of ethylene was below the detection limits of this study. Of the gases considered, ethylene's solubility in water is second only to carbon dioxide. One must conclude that any ethylene produced must be dissolved in the water column and must not appreciably participate in stripping or ebullition.

Low amounts of carbon dioxide were found in samplers set out after the 1st of September. The solubility of carbon dioxide in water is almost 20 times that of the other gases considered. Very high concentrations of carbon dioxide would thus have to be present for it to participate in stripping due to this high solubility in water. These high concentrations of carbon dioxide are possibly due to the oxidation of the peak levels of methane produced during the previous month or two (graph 3T, 2T).

The quantity and composition of gases produced in lake sediments could be used as an indicator of the trophic status as well as the level of organic degradation. Various hydrocarbons might even indicate the presence of specific microorganisms and processes (Porcella et al., 1975).

In conclusion, a marked yearly trend in gaseous ebullition has been shown. Some of the factors that influence this phenomenon have been investigated. Perhaps one of the most important lessons learned is the difficulty with which one collects reliable data concerning natural

phenomena.

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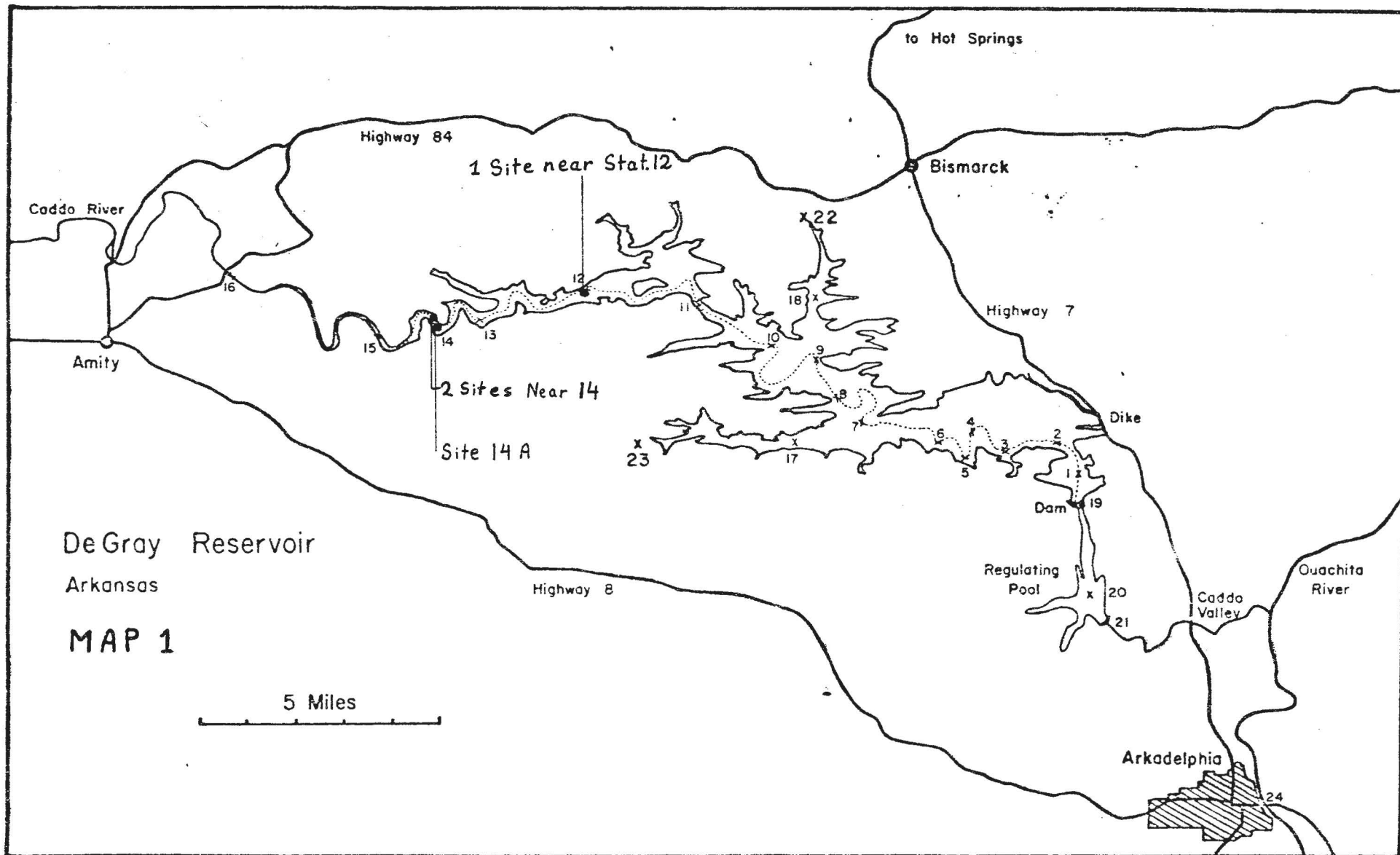
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Gas Collection Sampler

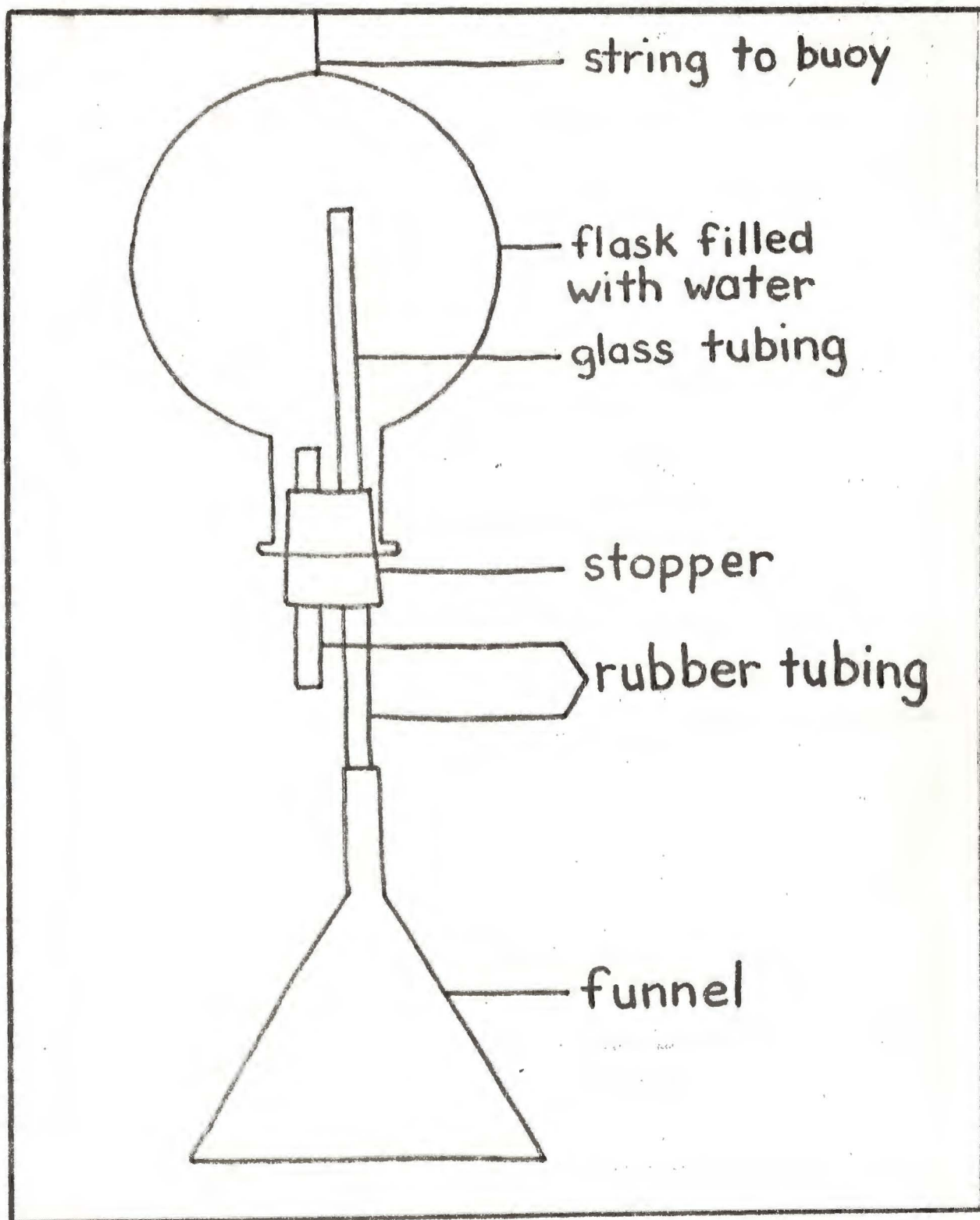
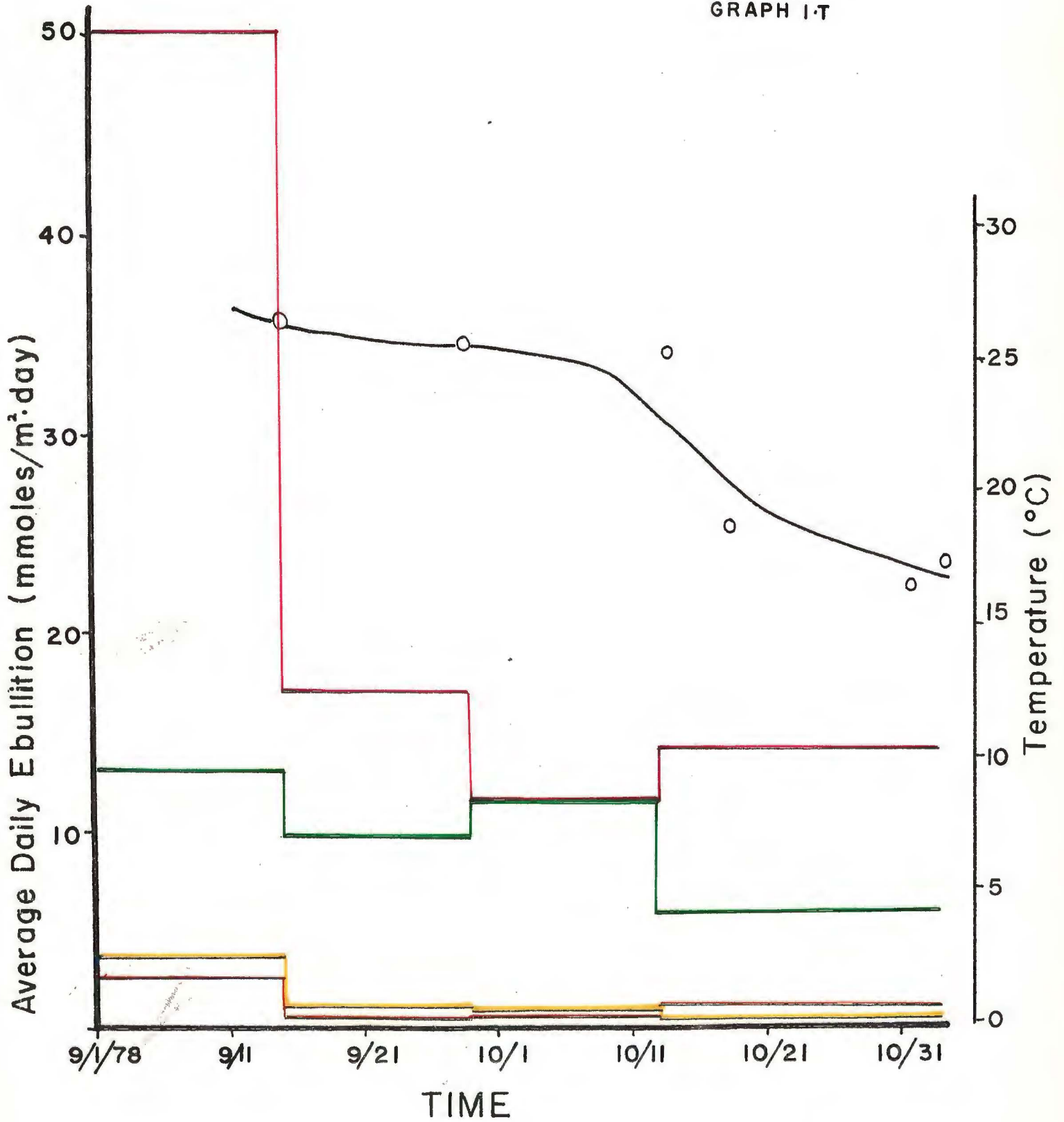


figure 1

MAP AND GRAPH LEGEND

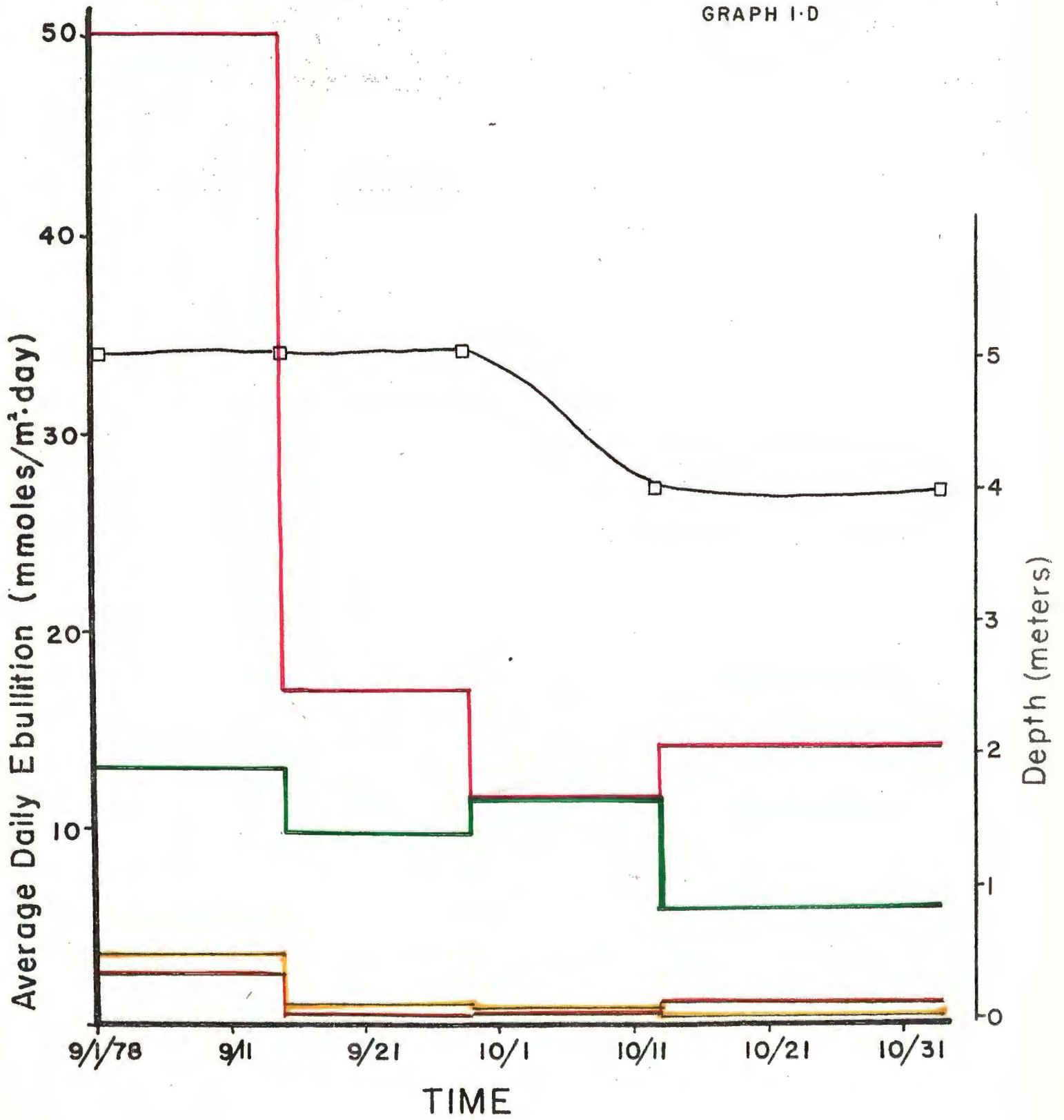
Temperature	—○—
Dissolved Oxygen	—△—
Depth	—□—
Nitrogen	—■—
Oxygen	—■—
Methane	—■—
Carbon Dioxide	—■—
Ethane	—■—

SITE 14A
GRAPH 1-T

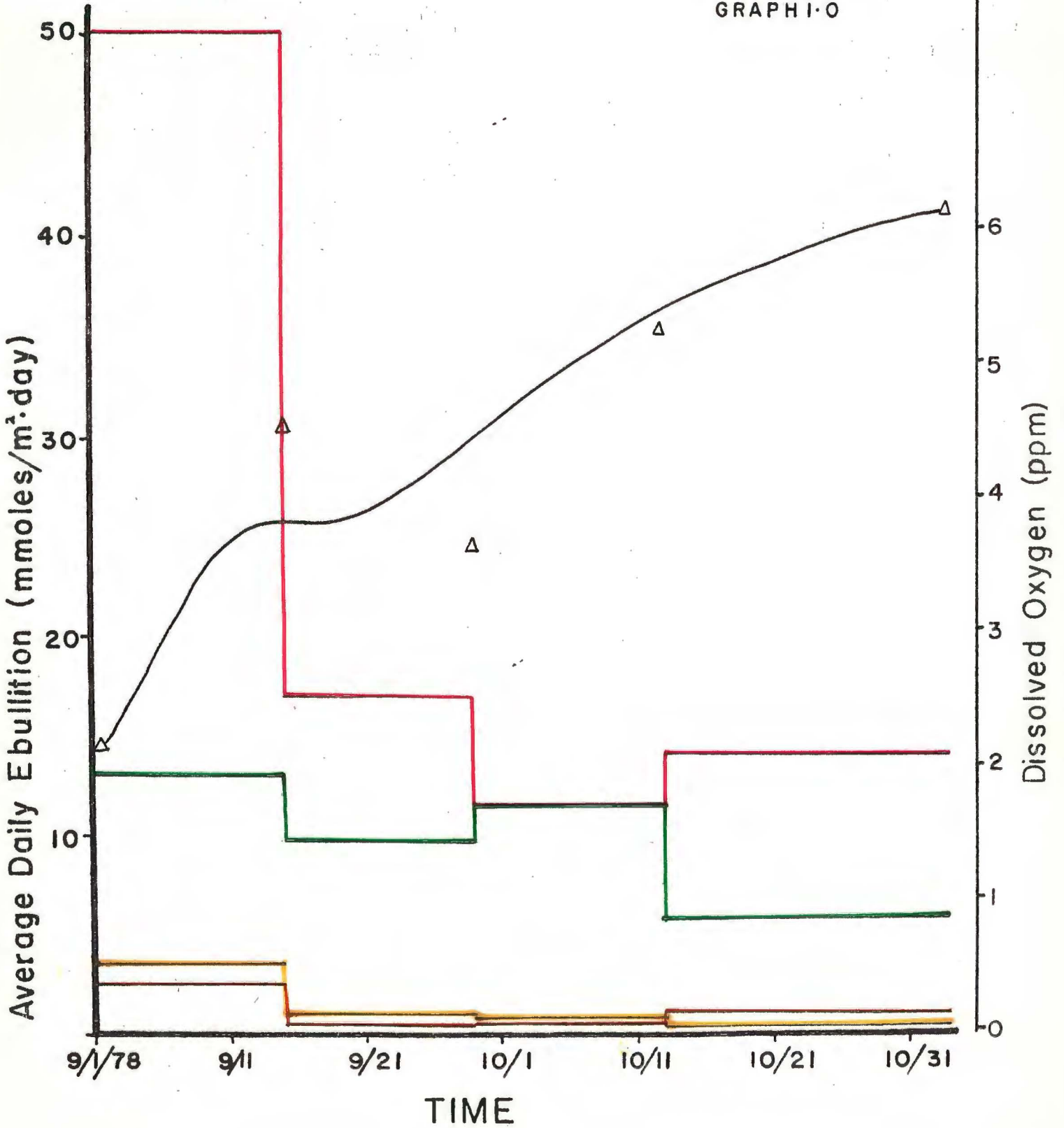


SITE 14A

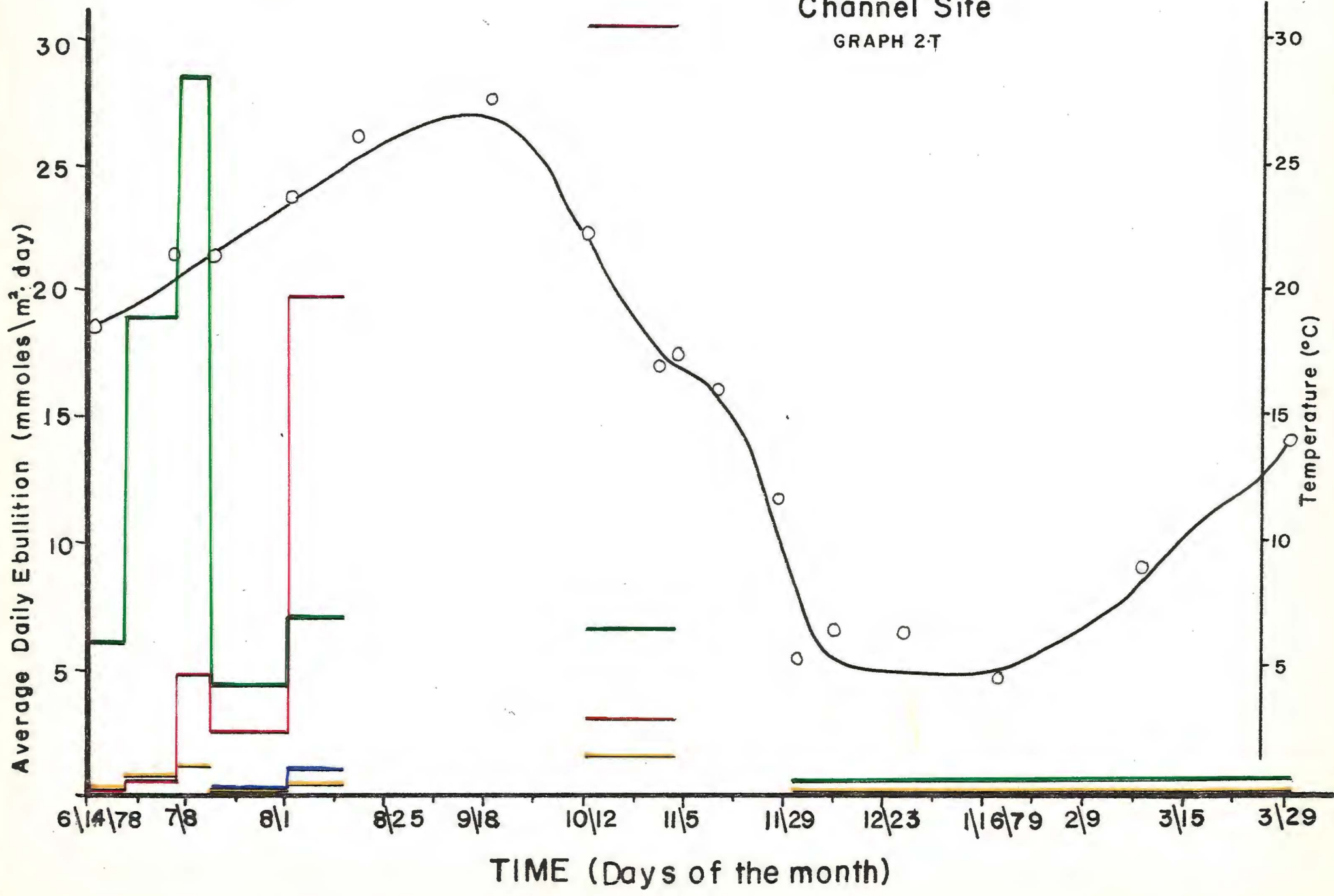
GRAPH 1-D



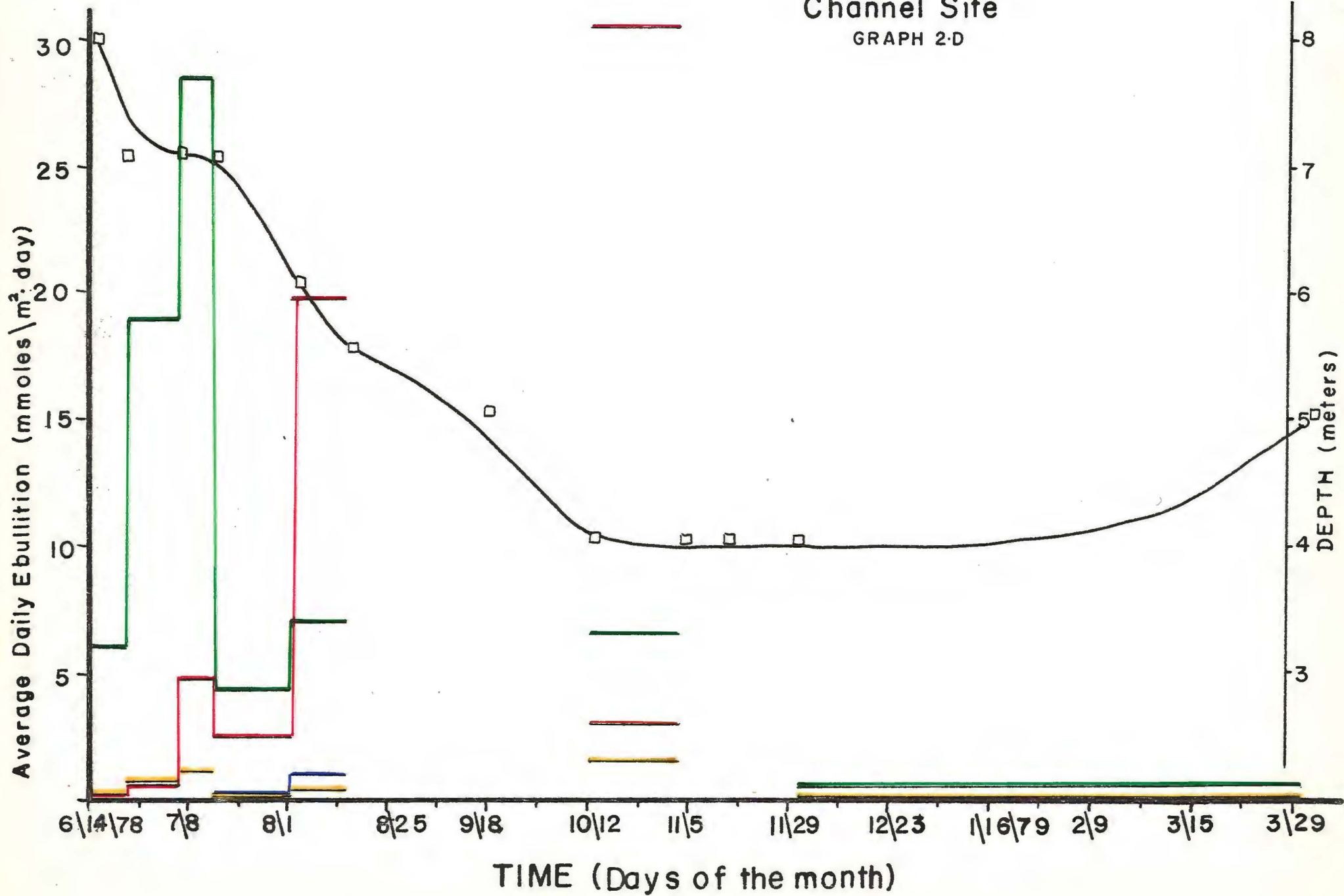
SITE 14A
GRAPH 1-0



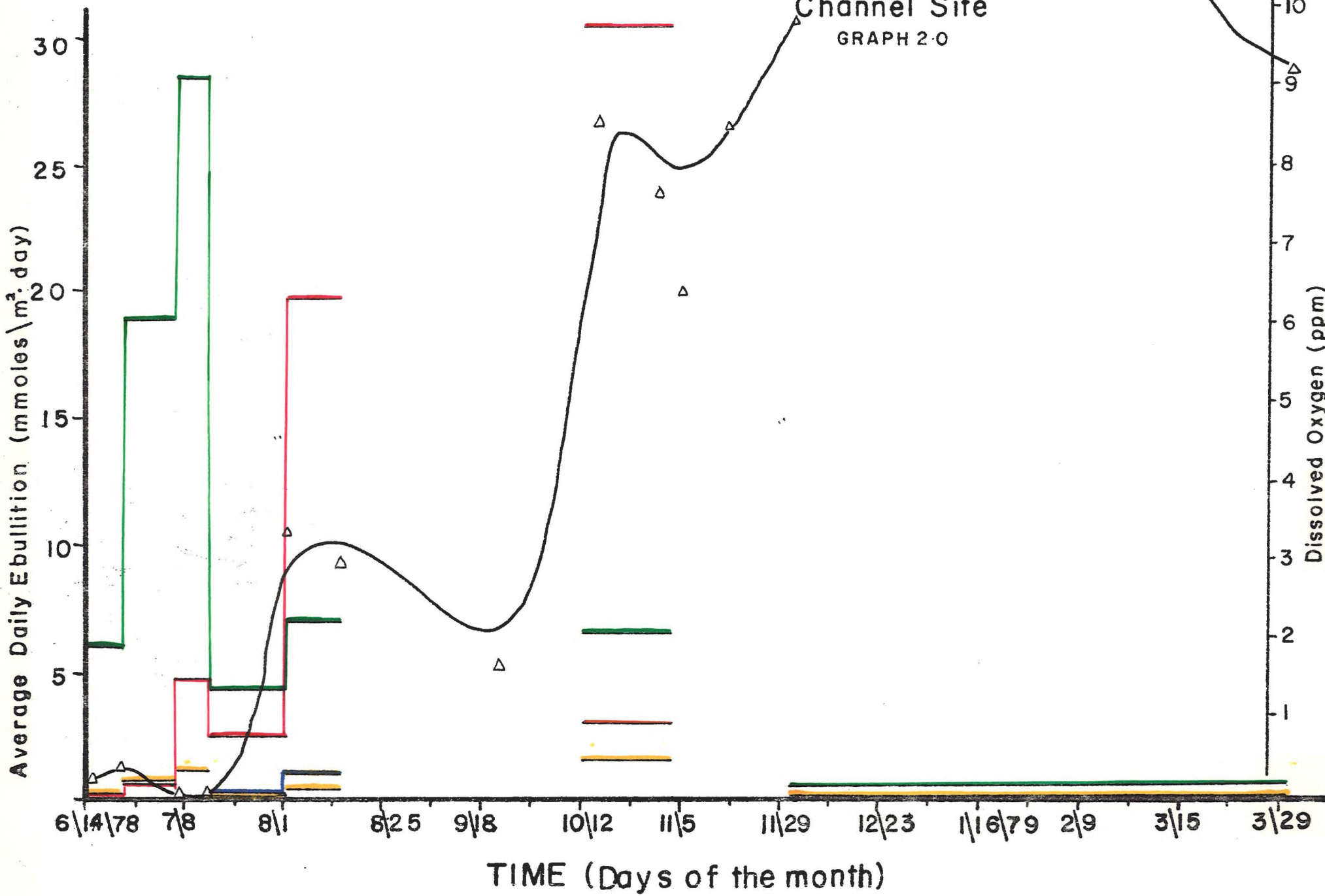
STAT. 14
Channel Site
GRAPH 2-T



STAT. 14
 Channel Site
 GRAPH 2-D



STAT. 14
Channel Site
GRAPH 2.0



METHODS

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In observing graphs 1T and 1D one sees that the dependence of methane ebullition on temperature and depth also holds for site 14A. Temperature data from the channel site at 14 (only 200m north of 14A and at the same depth) (graph 2T) is useful in determining that the temperature near the sediment water interface is actually falling during the sampling period in question.

Average methane ebullition over the channel site at

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Maximum measured methane ebullition at the creek mouth site near station 14 was more than three times greater than that over the channel site (98.0 mmole/m².day vs. 30.5 mmole/m².day). The greater activity over the creek mouth is possibly due to pre-impoundement plant life and post-impoundement sedimentation rates. This data, though, might be misleading due to differences in temperature and depth between the two sites.

Oxygen is present in small quantities in almost all the samples. Though some is possibly due to contamination during sample injection, most probably stems from stripping from the aerated water column. The quantity involved is lower than that of nitrogen due to oxygen's higher solubility in water.

An interesting phenomenon was the ebullition of ethane from the two sites at station 14 during the early stages of ebullition with the disappearance of ethane from the evolved gases after the sample taken on the 15th of August (graphs 2T and 3T). Due to the high activity at the creek mouth, one would expect relatively high amounts of ethane to be present (Smith et al., 1969). Its solubility, though, is even higher than that of oxygen. As the temperature of the sediments rises, and the solubility of ethane decreases, perhaps an initial excess of ethane is stripped as happened with nitrogen. After the initial removal of the excess ethane, its solubility is so high that it does not participate in much stripping.

Porcella et al. (1975) have recorded large amounts

of ethylene produced in their anaerobic microcosms. The ebullition of ethylene was below the detection limits of this study. Of the gases considered, ethylene's solubility in water is second only to carbon dioxide. One must conclude that any ethylene produced must be dissolved in the water column and must not appreciably participate in stripping or ebullition.

Low amounts of carbon dioxide were found in samplers set out after the 1st of September. The solubility of carbon dioxide in water is almost 20 times that of the other gases considered. Very high concentrations of carbon dioxide would thus have to be present for it to participate in stripping due to this high solubility in water. These high concentrations of carbon dioxide are possibly due to the oxidation of the peak levels of methane produced during the previous month or two (graph 3T, 2T).

The quantity and composition of gases produced in lake sediments could be used as an indicator of the trophic status as well as the level of organic degradation. Various hydrocarbons might even indicate the presence of specific microorganisms and processes (Porcella et al., 1975).

In conclusion, a marked yearly trend in gaseous ebullition has been shown. Some of the factors that influence this phenomenon have been investigated. Perhaps one of the most important lessons learned is the difficulty with which one collects reliable data concerning natural

phenomena.

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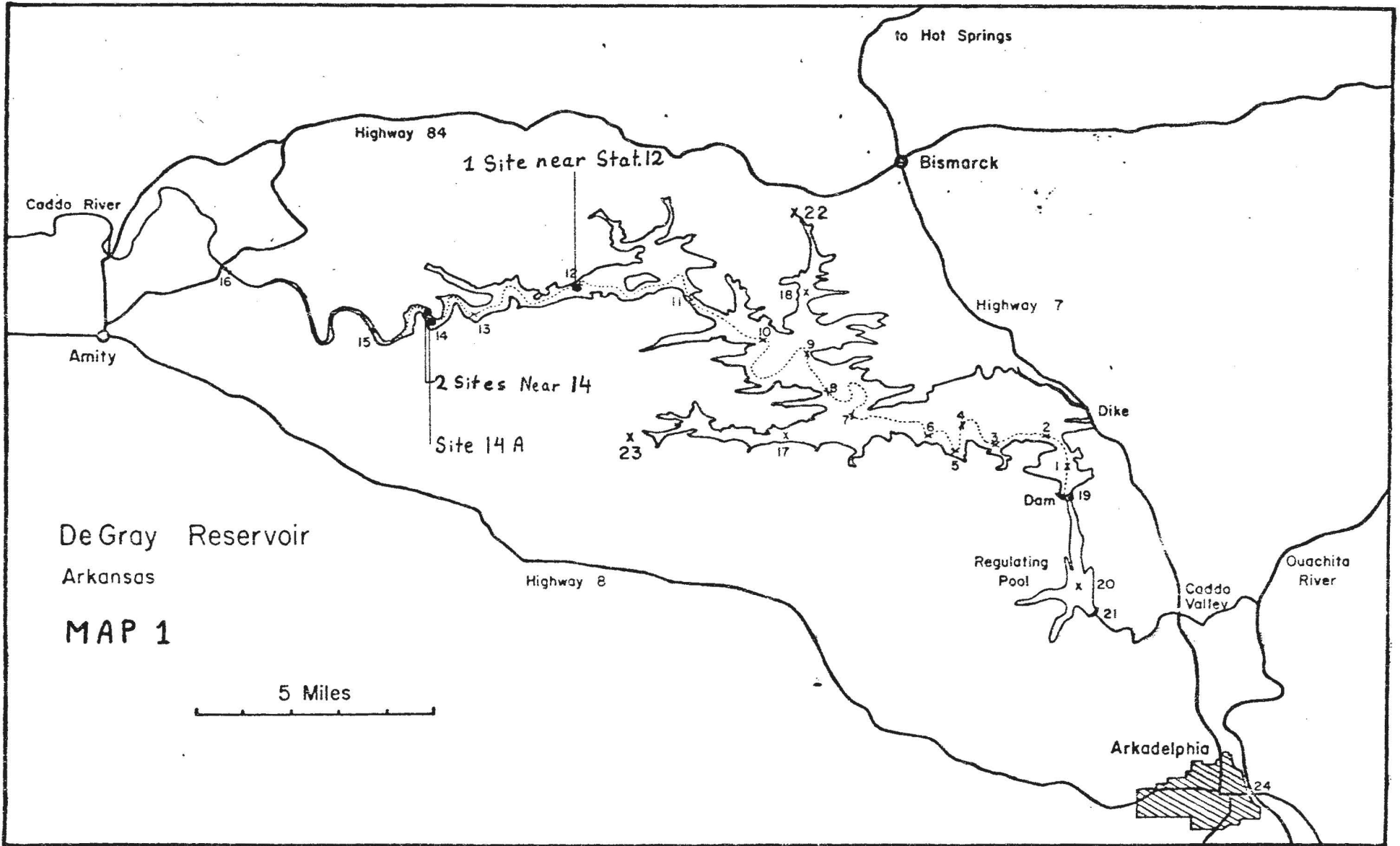
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Gas Collection Sampler

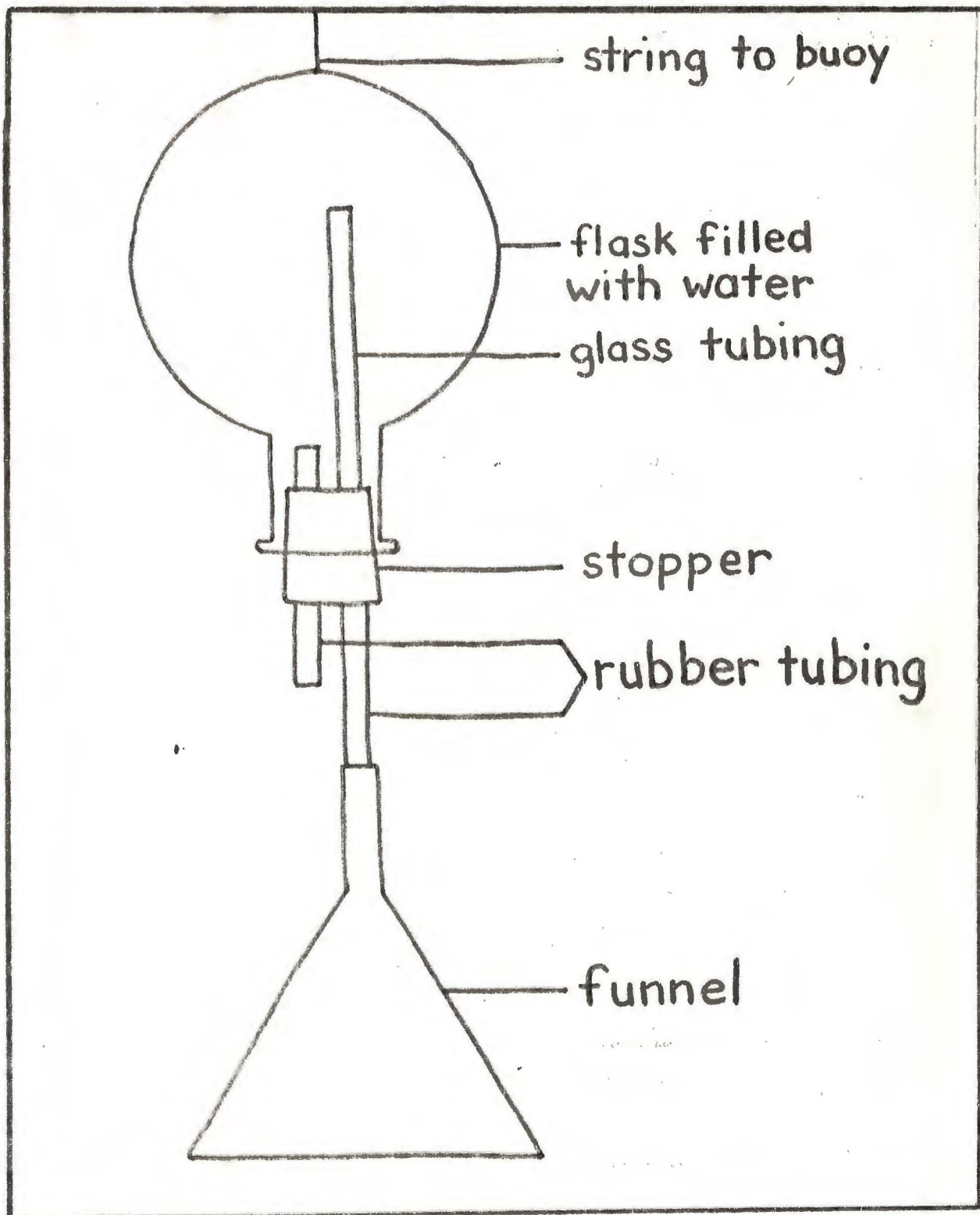
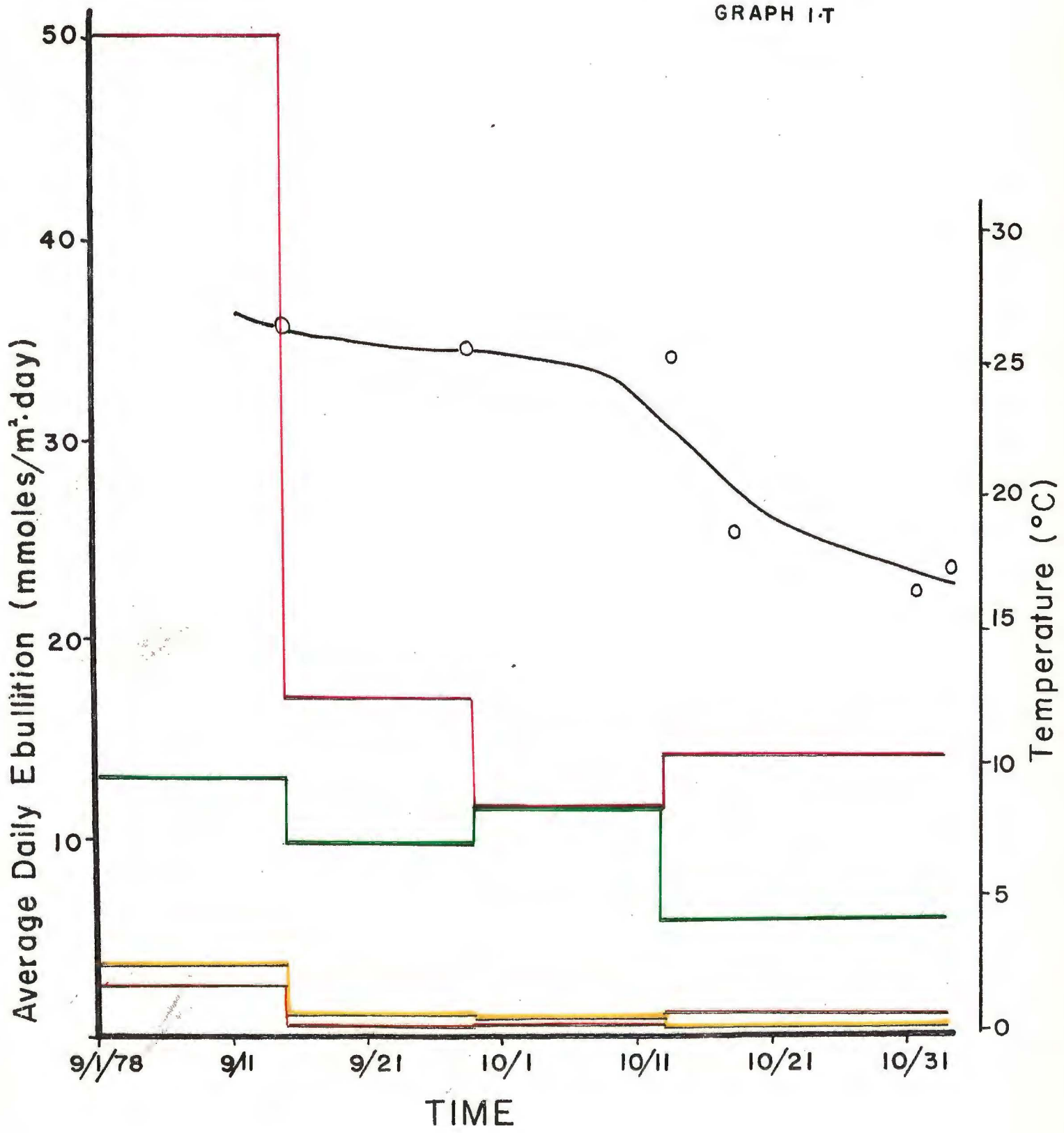


figure 1

MAP AND GRAPH LEGEND

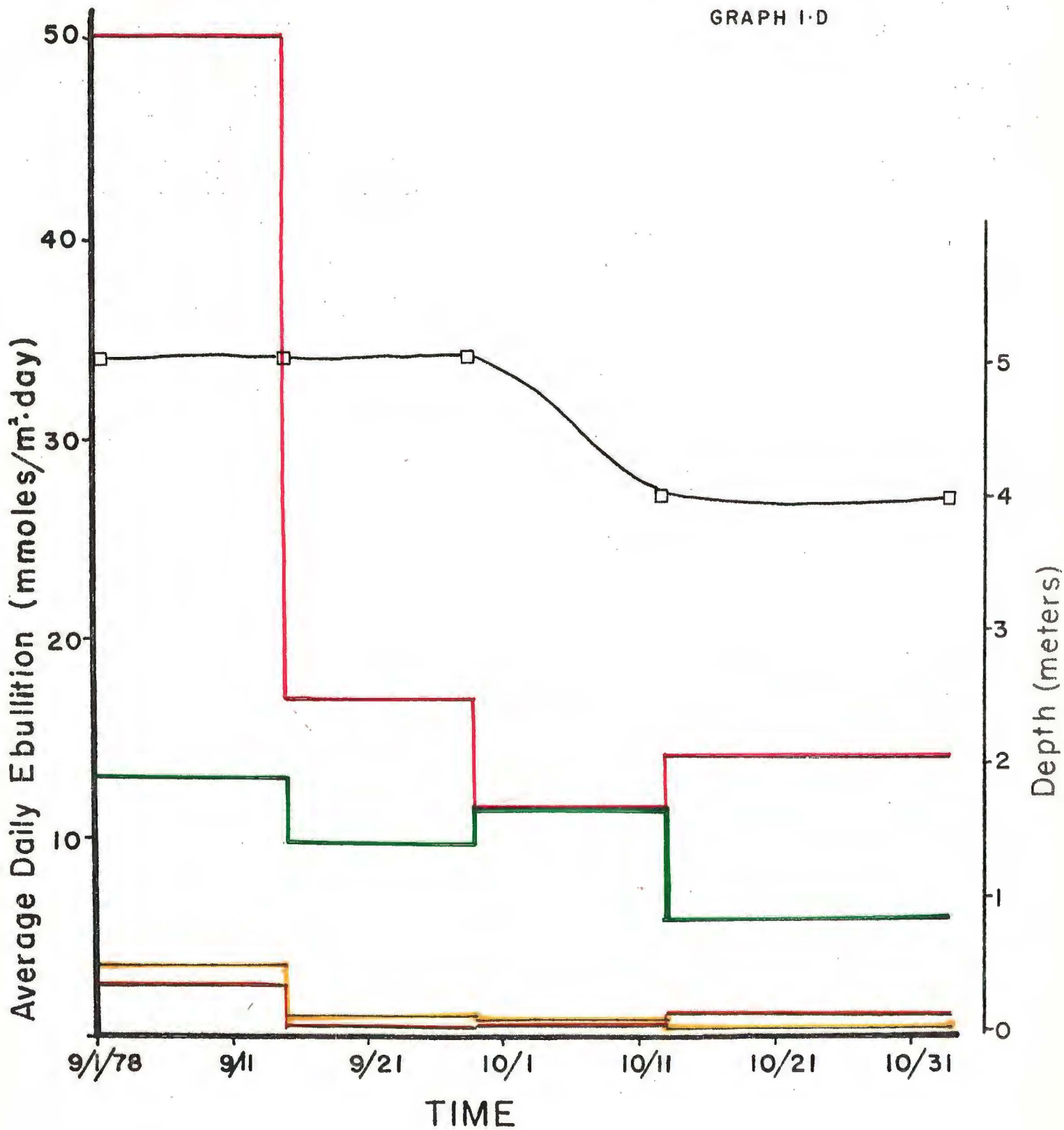
Temperature	—○—
Dissolved Oxygen	—△—
Depth	—□—
Nitrogen	—
Oxygen	—
Methane	—
Carbon Dioxide	—
Ethane	—

SITE 14A
GRAPH 1-T

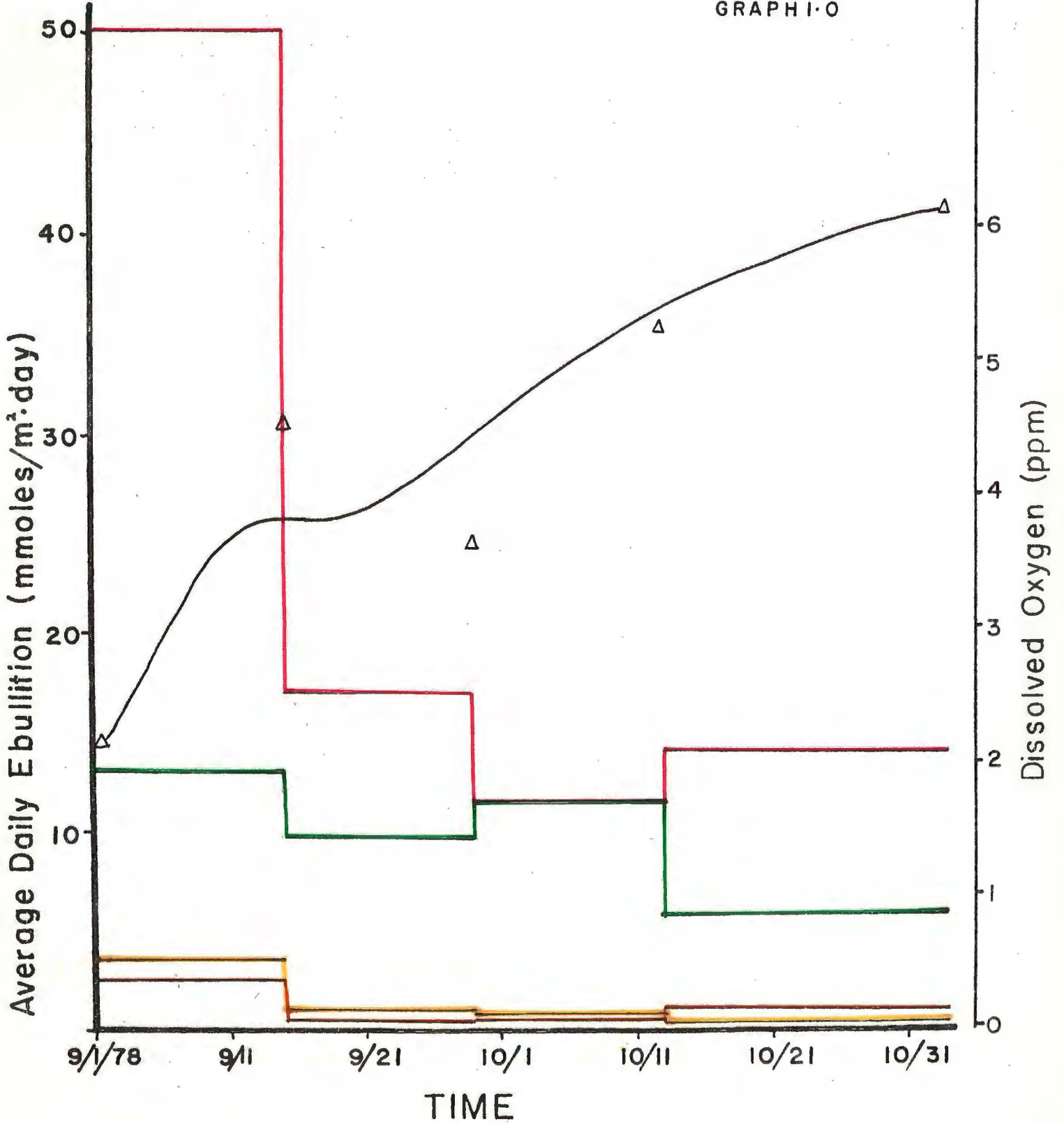


SITE 14A

GRAPH 1-D

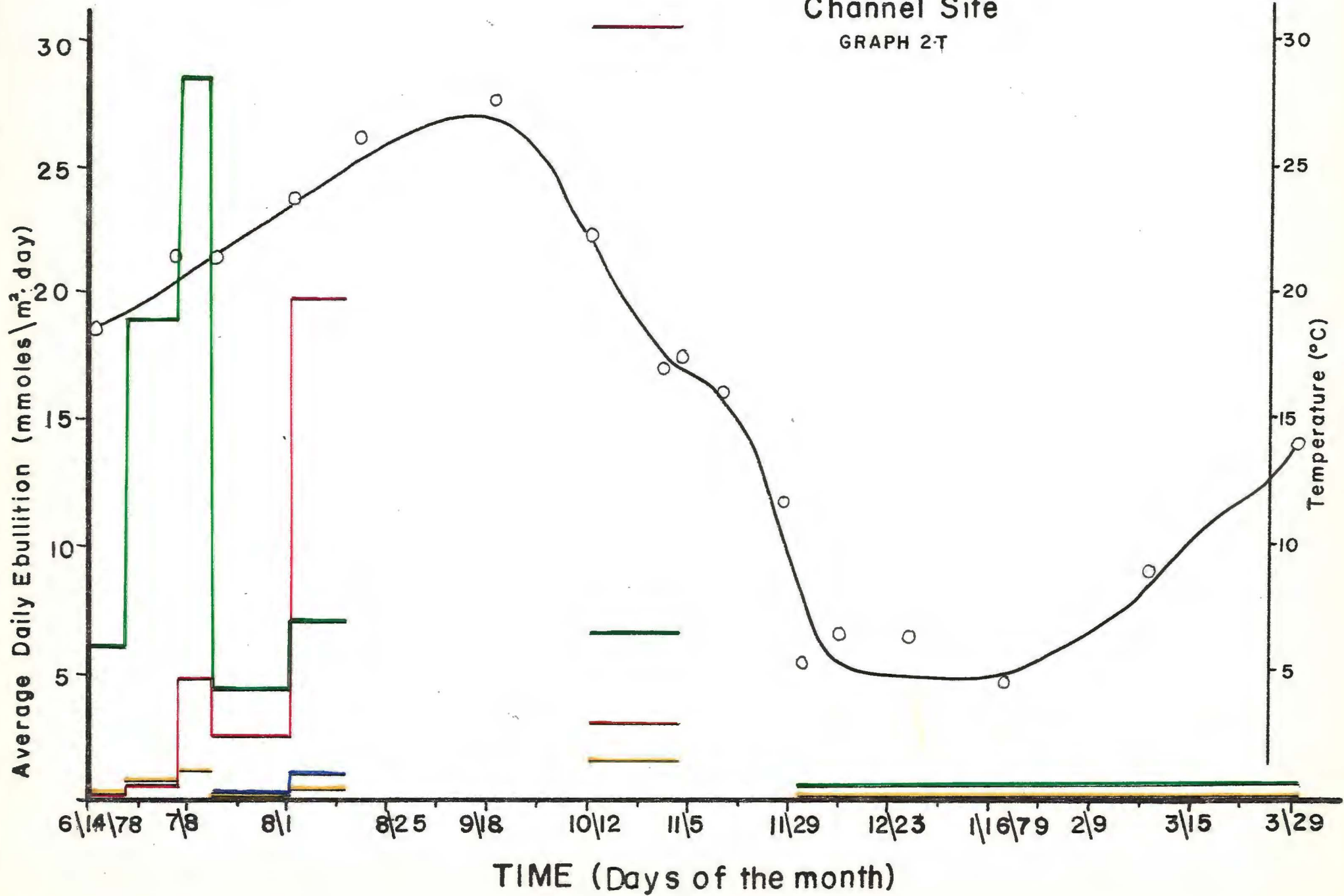


SITE 14A
GRAPH 1.0

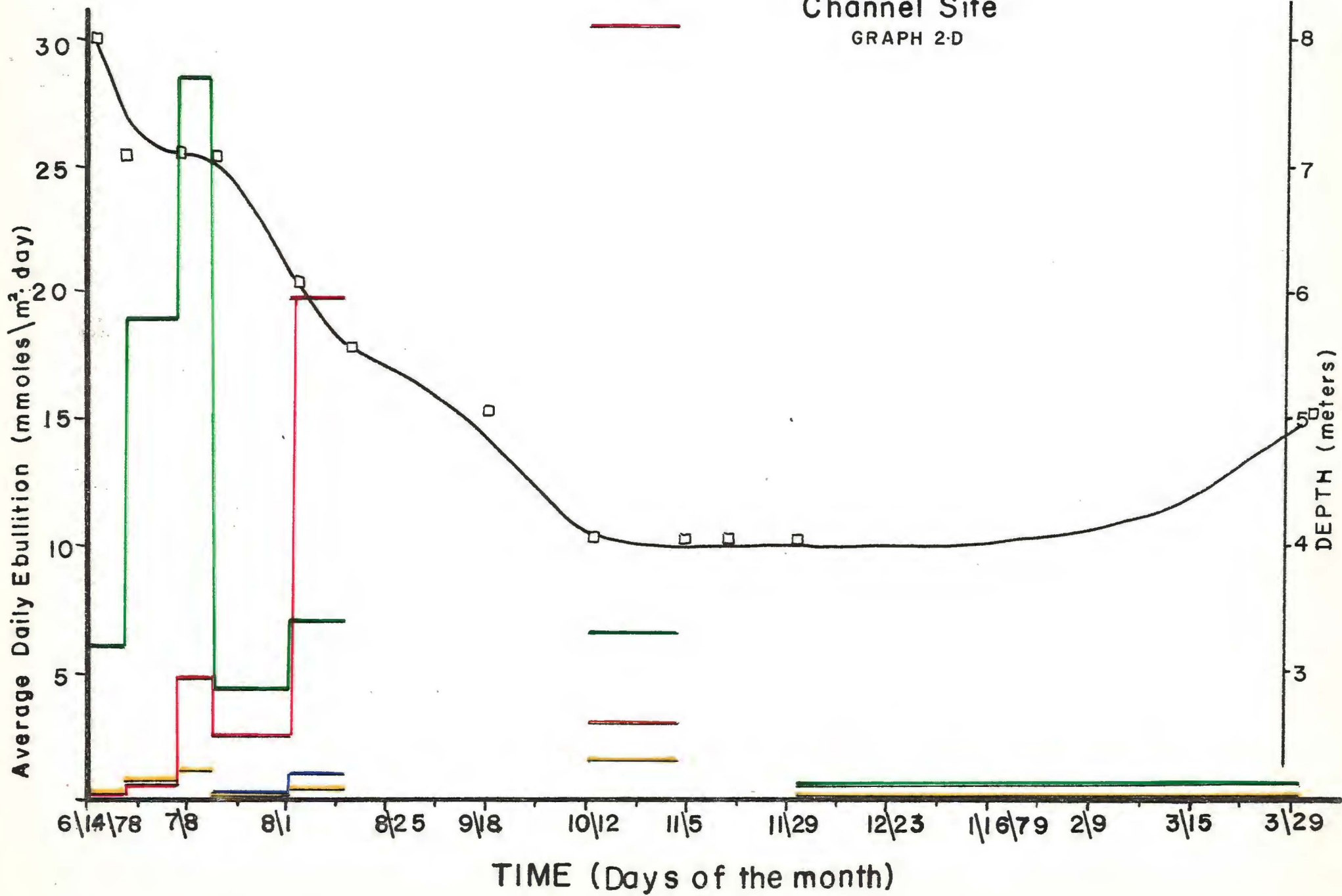


STAT. 14
Channel Site

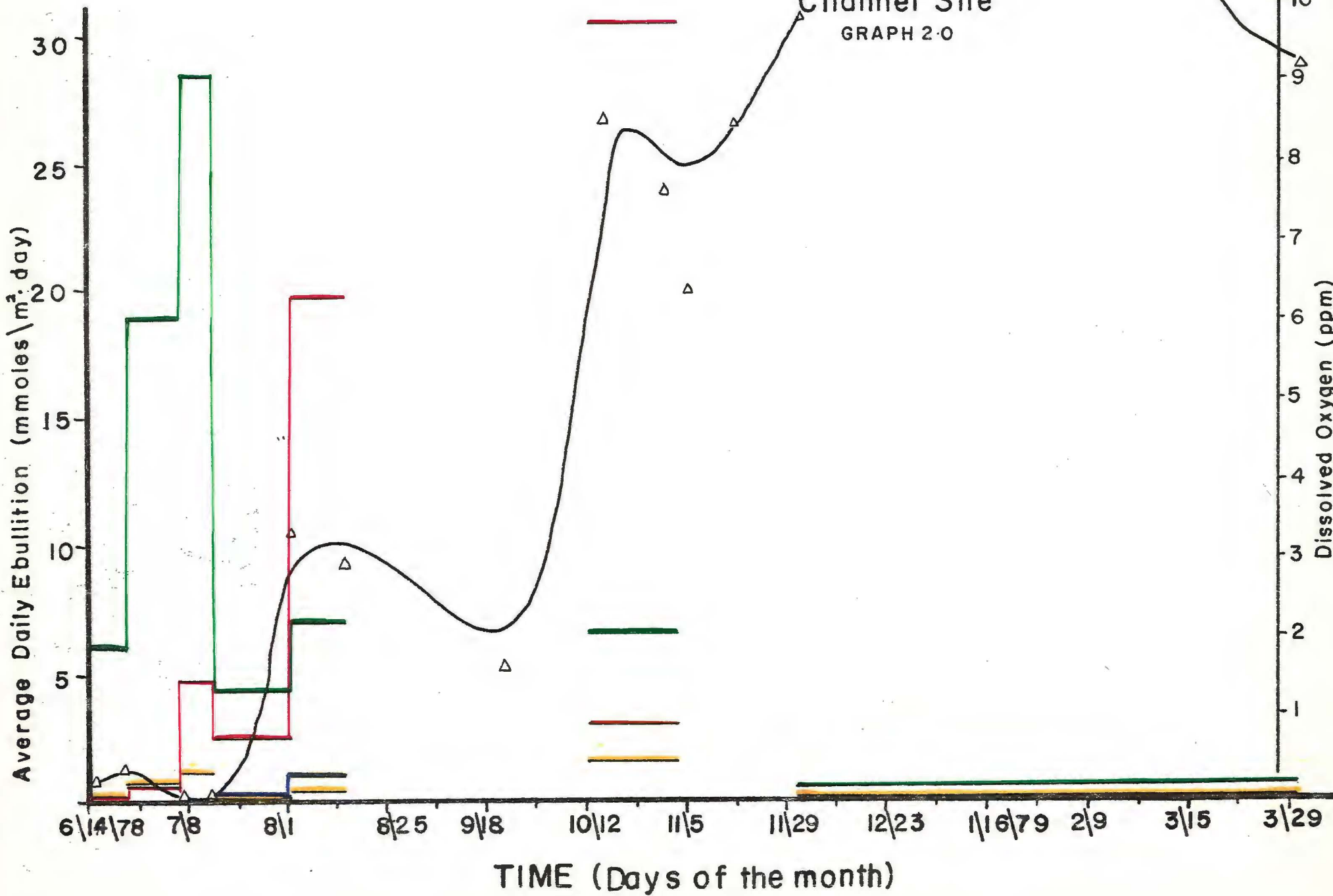
GRAPH 2-T



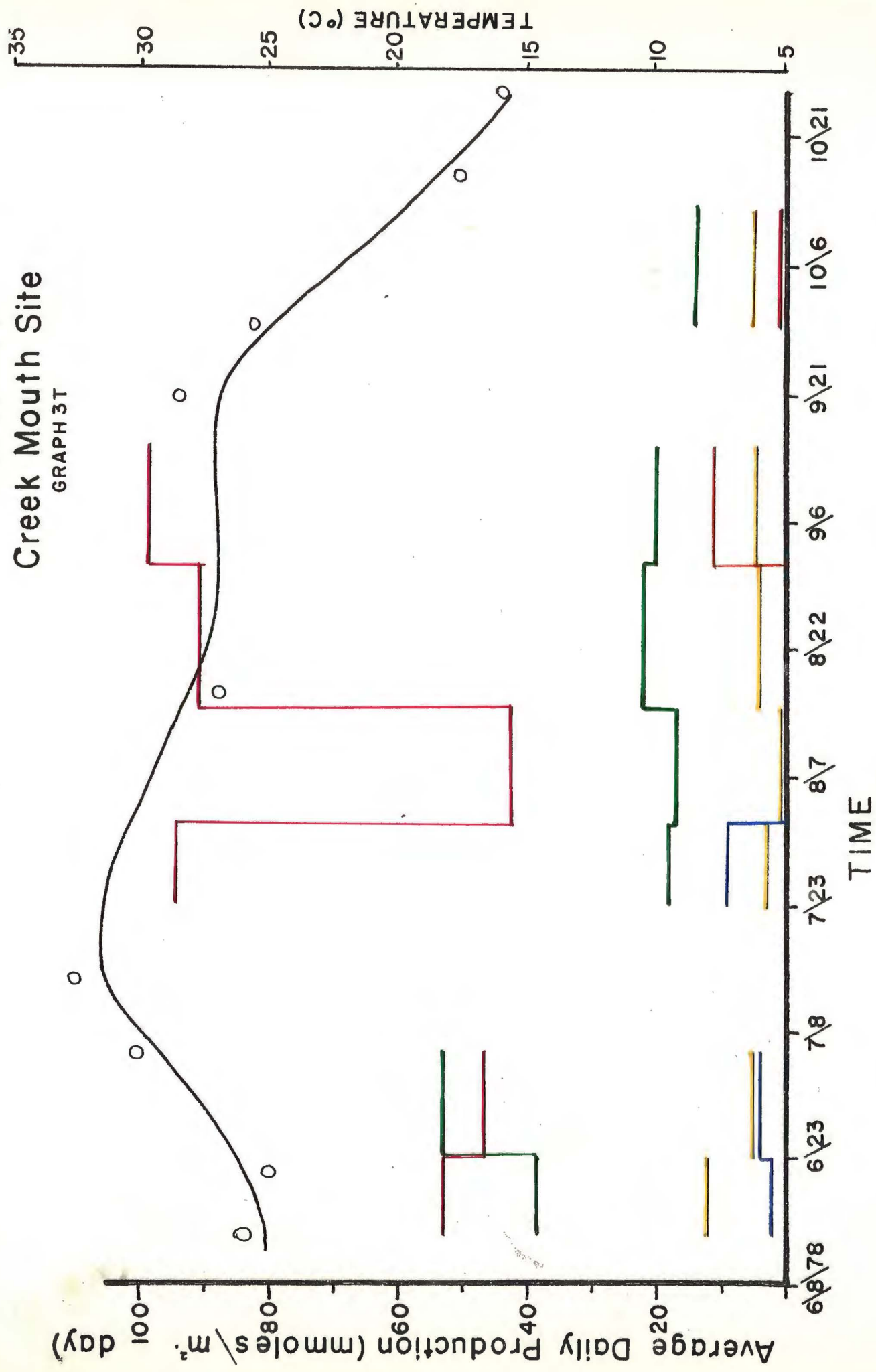
STAT. 14
 Channel Site
 GRAPH 2-D



STAT. 14
Channel Site
GRAPH 2.0

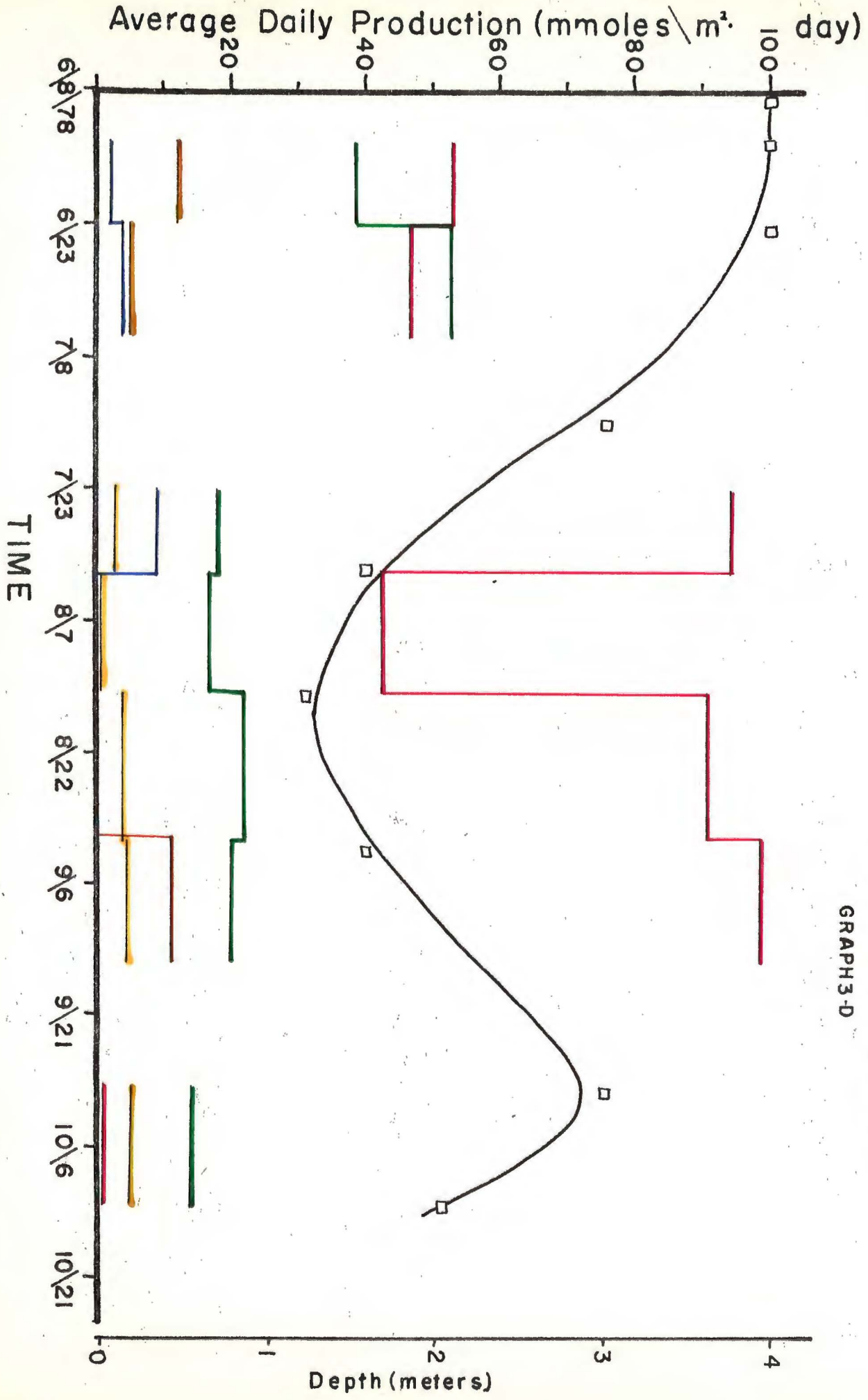


STAT. 14 Creek Mouth Site GRAPH 3T



STAT. 14
Creek Mouth Site

GRAPH 3-D



STAT. 14
Creek Mouth Site

GRAPH 3.0

