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HYDROCARBONS FROM PLANTS AND TREES

Melvin Calvin

July 1982

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HYDROCARBONS FROM PLANTS AND TREES

Melvin Calvin

Department of Chemistry and Lawrence Berkeley Laboratory
University of California, Berkeley, California 94720

Plenary Lecture, Symposium on Biomass Substitutes for
Liquid Fuels, Campinas, Brazil, February 10, 1982

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Introduction

Professor Monaco suggested that we (Mrs. Calvin and I) were in some way helping to solve Brazil's problems. Actually, Brazil's problems are the world's problems and we are concerned with world problems. If Brazil happens to have problems which are similar to world problems, then we can help there also. We have been to Brazil a number of times and have had occasion to visit a good part of the country. In actuality, in our conversations with various friends here we have discovered that most of the people we meet who are native to Brazil have not been to as many different parts of the country as Mrs. Calvin and I have been in the last eight years. This allows us, perhaps, to show you some bits of Brazil that even Brazilians have not seen.

The topic of this meeting is the possibility of obtaining oil from green plants. As a means of introducing this topic it would be useful to examine the way in which energy was used in the United States (a developed country) in 1980. This diagram shows the development of energy from both its source to its end use, and how the energy is converted from its primary source to its final form (Fig. 1). There are three messages contained here:

- (1) The first is that oil, natural gas and coal constitute 74 out of 78 total units of energy used in the United States in 1980. These three components consist almost entirely of ancient photosynthetic products made several hundred million years ago by the action of sun on the green plants, stored

in various forms in the earth and gradually converted into the forms we now know--oil, gas and coal. The first message, therefore, is that we in the United States (and, of course, throughout the world) are heavily dependent on the fossil photosynthetic products.

(2) The second message concerns the way in which energy is used. When the energy contained in these fossil forms is used, it is almost always used by first burning the material with air, thus converting the chemical energy to heat and using the heat in various ways. Generally that heat is converted into mechanical work, when electricity is generated, when turbines of an airplane are being turned, or when an automobile is being driven, for example. There is a limitation on how much mechanical work can be obtained from that energy, the well known Carnot limitation, and as a result of that law an enormous amount of the primary chemical energy is lost as waste heat in conversion and transmission in mechanical engines. For example, in transportation, from 18 energy units input, 16 is lost as low grade heat. Only when that primary energy is used directly as residential or commercial heating is the loss a small one. The relatively high grade energy has to be first converted into heat and then into work, which constitutes a relatively inefficient use. The reason I emphasize this point is that later on when we discuss alternatives to these fossil sources, one of the alternatives will be to generate heat and that route is always affected by the Carnot limitation.

(3) The third message concerns predictions about diminishing oil supply. When that subject arises, even today, you often hear the response that actually there is no problem with the supply of petroleum and always some new oil has been found. I want to indicate that I feel there is a limit on how much oil will be available. So far as we know, this material is the product of ancient photosynthesis and therefore is present in a finite amount. It is hard to estimate actually how much oil is available in the earth's crust with any kind of precision, but this can be done crudely in several ways. The economists use price as a criteria, with the idea that an increased price will mean increased oil supply. However, the price of oil has in it other factors than geological availability--social and political components, as we have learned. I sought to find another way to estimate the true availability of oil. Someone did this recently, using the existing history of oil discovery from 1945-1975 (Fig. 2) which shows that the number of barrels of oil found per foot of well drilled is falling. Earlier, King-Hubbert prepared a similar type of curve. From these two pieces of information it is possible to see that in 1945 it was possible to obtain 15 barrels per foot of well drilled and in 1975 it was necessary to drill twice as far to find a barrel of oil. This shows that the effort (energy) required to find a barrel of oil has doubled in the last 30 years. This information can be converted into energy cost for drilling and extracting the oil and that cost is constantly rising. Therefore, the

energy costs to find a barrel of oil are constantly increasing.¹ The information in Fig. 2 shows that about the year 2000 the energy costs to find a barrel of oil and get it from the ground will exceed the energy content of that barrel of oil. Thus, even if there is oil available, it might not be practical to use up more energy to find and extract it than is represented by the barrel of oil itself.

There are other fossil resources, of course, and there is an historic pattern as to the way energy sources come into use, i.e., an energy source is used up to its peak and gradually disappears as another energy source comes in. King-Hubbert predicted that the curve for oil would peak somewhere between 1980 and 2000, and I believe that is what is happening in terms of productivity. However, when that same type of curve was constructed for coal it appeared that the coal would peak later, indicating that coal at least in the United States will last several hundred years. This has led, in our country (and in Germany, Great Britain and the Soviet Union as well) to the idea that if coal is available it can be converted into a form more appropriate for our needs, i.e., liquid, and can become the fossil fuel for the next decades.

Carbon Dioxide Problem -- The Greenhouse Effect

There is a limitation, however, on the use of coal. As you may recall, some twenty to thirty years ago in our country we were persuaded to convert our steam plants from coal burning to clean oil or gas burning plants, which was done. Now, the

persuasion is to return to coal. The reasons we left coal in the first place are still with us, that is, the carcinogen in the ash, the acid, the sulfur, etc. We can clean up the ash, carcinogens, sulfur, and nitrogen oxides for a price. But, there is one factor that cannot be avoided if coal is to be used as a cheap source of energy, and that is the production of carbon dioxide. The amount of CO_2 produced per unit of energy generated from coal plants is roughly twice as much as it is from any other resource. That is the primary reason for being circumspect about expanding the use of coal.

The historical record of the carbon dioxide in the atmosphere from 1958-1981 has been plotted (Fig. 3) and it is clear that each winter the CO_2 rises (with increased burning of coal throughout the world for heat) and falls each summer. However, each year there is some carbon dioxide left over, with the net result that the CO_2 level in the atmosphere is rising. During that period (1958-1981) the CO_2 concentration has risen by about 5% and when we go back to 1858 it is possible to estimate that the carbon dioxide content of the atmosphere was much less. This can be done by an isotopic measurement of the carbon-13 and carbon-14 content of tree rings that were laid down one hundred years ago. The value for the CO_2 concentration has gone from 290 ppm in 1858 to 330 in one hundred years, about 15% in the last century and 5% in the last twenty years. The CO_2 concentration is rising much more rapidly now than it was one hundred years ago.

There is one physical property of the carbon dioxide which makes this rise in concentration very important and this is known by its consequence as the "greenhouse effect" (Fig. 4). The CO₂ blanket is transparent to the visible light of the sun. Wherever the light strikes the earth's surface it must ultimately be converted to heat. Carbon dioxide is opaque to infrared light; it absorbs that infrared light and re-reflects it back, acting as a one-way valve for energy from the sun, allowing it to come in to the earth's atmosphere and allowing only a very small fraction to escape. The ultimate consequence of this effect is a warming trend in the earth's climate.

Efforts have been made (and are being made) to detect that warming trend which might have occurred as a result of the 15% rise in CO₂ concentration in the last one hundred years.² The annual fluctuations of climate are so great that the rise of temperature is still in the "noise" of these fluctuations, and it is a matter of sophisticated computer analysis to sort out that particular effect from other effects. That's why it is not really easy to say that the earth has already begun to feel the warming trend of that increase in CO₂ concentration.³ My personal feeling is that if we wait until the temperature rise becomes large enough so that it can be seen easily among the annual fluctuations, it will be too late. It is necessary to consider this problem now.

One way to help control the increase of carbon dioxide in the atmosphere is to limit how much coal is introduced into the

energy system. When various methods of using coal as an alternative to oil (coal gasification, coal liquifaction, etc.) become more prevalent, the problem of increased CO₂ concentration will become one of the larger factors to be considered. If energy is to be obtained from coal, CO₂ will be the result, and our use of coal must be limited in time and in amount, especially time, until other alternative sources of energy are found.

Sugar Cane as an Energy Source

We have been living on our energy capital, so to speak, our accumulated savings account of fossil carbon, accumulated over several hundreds of million years. It is quite obvious that we are putting back much of the carbon into the atmosphere that the green plants took out of it about 250 million years ago. We must now learn to live on our current income and not be completely dependent upon our accumulated capital. That current income, of course, is what the green plants fix every year in the form of reduced carbon.

Here in Brazil you have already taken steps in that direction on a substantial scale, through the alcohol from sugar cane program. In 1974 the production was 400 million liters of alcohol, mostly from molasses, and the 1981 production was 4.4 billion liters of fermentation alcohol, a tenfold increase. That is a real accomplishment and an example to the rest of the world. I think the alcohol which can be produced here may ultimately be more valuable as a chemical raw material than as a fuel, but that is another consideration.

In Brazil, as well as in other countries that grow sugar cane, the development work went to finding a cane that had a higher percentage of sugar, so that when a ton of cane was processed more sugar could be obtained. However, if you are interested in the total amount of fixed carbon, that may not necessarily be the only way. There is an effort now underway of taking some of the old clones that were discarded in the days of the search for higher sugar content for the cane and make use of them for what the Puerto Ricans have called "energy" cane (Fig. 5).⁴ The energy cane produces about 100 tons/acre (250 tons/hectare) of total biomass. The sugar content of this cane is low. However, if you calculate the sugar content per hectare, it is the same for both the "energy" cane and ordinary cane. The "energy" cane has three times as much total produce which is good for various purposes. The Puerto Ricans are planning to use some of the material from the "energy" cane to fire their power boilers to produce electricity on the South Coast of Puerto Rico. This is an experiment that bears watching.

Hydrocarbon-Producing Plants

There are some plants that take carbon dioxide very much further down on the reduction scale than carbohydrate. Sugar cane starts with carbon dioxide, a carbon atom with two oxygen atoms, and stores most of the captured solar energy in half-reduced carbon, a carbon reduced with one hydrogen atom and still bearing one oxygen atom. What we really want is fully reduced carbon, carbon with no oxygen and just two hydrogen

atoms, as in petroleum. Are there any plants that go further than carbohydrate? The answer is positive. In fact, the most well known of all, rubber (Hevea brasiliensis) was grown here in Brazil. We came to Brazil the first time in 1974 to see if there were any other plants of that same family which produced hydrocarbon of lower molecular weight than rubber and more of it.

We first examined rubber production in Malaysia when we visited there in 1975. We learned that until 1945 the Malaysian rubber trees produced about 200 lbs of rubber (hydrocarbon) per acre per year. However, the advent of World War II brought the development of synthetic rubber from petroleum. After the war, the Malaysians saw that their dominance of the market for natural rubber had disappeared and they had to improve the rubber yield in order to be competitive. Within the period of 1945-1965 the productivity of the rubber trees was multiplied by a factor of ten. By 1965, 2000 lbs of rubber per acre per year was practical and there were some experimental plantations that produced 4000 lbs of rubber per acre per year and individual trees which, if they could be planted at high enough density, could produce 8000 lbs of rubber per acre per year. This improved productivity was accomplished by the usual methods of plant breeding and plant selection on a species that had been grown commercially for over one hundred years in Brazil and Malaysia.

We examined plants as candidates for oil production and the first one we chose belonged to the same family as the rubber tree,

the family Euphorbiaceae.⁵ There are over 2000 species of Euphorbias, of many sizes and shapes growing throughout the world. They all have one property in common, that is, they secrete a latex which is made up roughly of one-third hydrocarbon and a few percent protein and a small percentage of carbohydrates. It is the hydrocarbon in the latex that would be the product of interest. The various candidates are characteristic for certain areas, growth habits, maturation times, harvestability, etc.

Our first candidate for study, Euphorbia lathyris (Fig. 6) was studied extensively in California and in the semiarid area in Arizona. We wanted to find a plant that would grow on land not suitable for food or fiber production, and the agronomic experimental plantations developed since 1977 have provided a great deal of new information on this species. Plantations of E. lathyris are also under cultivation in Spain (Fig. 7) under the sponsorship of the Ministry of Energy and Agriculture, and studies are underway in Australia as well to see whether or not this species can be adapted to their climatic and soil conditions.⁸

In the Canary Islands, for example, there are three other species (E. Regis Jubae, E. balsamifera and E. Canariensis) (Fig. 8) which are suitable candidates. The latex from E. balsamifera is called a "sweet" latex and is used as cream in drinks such as tea by the people who live there. From what we know, it is predicted that the Spanish will use the

E. balsamifera as well as E. lathyris for an energy plant under their climatic conditions. In Puerto Rico we found another species, E. lactea (Fig. 9) which grows in profusion on the dry south coast; when the trunk of the E. lactea is cut with a knife, a profuse flow of white latex occurs. Still another species which grows extensively in Brazil is Euphorbia tirucalli which is used as a fence to keep cattle from the cane fields; plantations of this same species are under cultivation for oil in Okinawa (Fig. 10).

The material from the E. lathyris produces not only oil but carbohydrates (fermentable sugars) as well, making it an energy efficient candidate for alternative agriculture. We do not bleed the plant for its latex but cut the plant, let it dry in the field, pick up the dried plant, extract it with various solvents. From 100 dry tons of E. lathyris the yield is 8 tons of oil. Much to our surprise, after the oil was extracted with hexane, we learned it would be possible to extract sugars with methanol-water mixtures (Fig. 11). This route produced 200 tons of fermentable sugars to produce alcohol. This was a surprising development, because when we started this work we concentrated mostly on oil production and did not realize that sugars (alcohol) would also result. The Euphorbia lathyris produces approximately equal volumes of sugar-alcohol and oil.

As mentioned earlier, the latex of the Euphorbias is the product in which we are interested. Most of the Euphorbia

latexes have an irritant in them, usually a phorbol ester, which irritates the skin and can cause blindness if it gets into the eyes. When the material from E. lathyris is harvested, the toxic substance practically disappears as the result of drying, and by the time the material is extracted by solvents about 99% of that remaining is eliminated. Therefore, Euphorbia lathyris does not present a particularly toxic material for an alternative energy crop.¹⁰

Here in Brazil we found another family, Asclepiadaceae, which also contains many different species which would be good energy agriculture candidates. One example (Fig. 12) is Calotropis procera which contains a latex similar to that in E. lathyris in chemical composition. Milkweed are common also in Australia and there are two rather large scale projects underway using C. procera as a source of oil and sugar in northern and eastern Australia.¹¹

Hydrocarbon-Producing Trees

When we visited Brazil in 1978 we heard, at a meeting in Fortaleza, about another candidate for hydrocarbon production that was actually a tree. It wasn't until we were able to visit the Ducke Forest near Manaus and see it for ourselves that the potential of this tree to supply oil for energy became apparent. This oil-producing tree belongs to the genus Copaifera, of the family Leguminosae, and produces terpenes. The product of the Copaifera multijuga, for example, is a mixture of sesquiterpenes, an oil which has the proper chemical

composition for use directly in diesel engines. This particular tree (Fig. 13) is harvested by drilling a hole in the trunk about 3 ft from the ground; the hole is about 2 cm in diameter and goes into the heartwood of the tree. A pipe is inserted in the hole and the oil drains out of the pipe into a bucket. This operation can be done twice each year and in 24 hours about 20 liters of material, similar to diesel fuel, accumulates. The hole is then plugged with a wooden bung and 6 months later the tree will produce another 20 liters from the same hole. The oil comes not from the cambium, as does the rubber latex in the Hevea brasiliensis, but from the heartwood, from 1-2 mm diameter pores running vertically throughout the trunk of the tree. There are at least 25 different compounds in the oil from these trees which have been analyzed by gas-liquid chromatography and each compound is a sesquiterpene.⁶

An experimental plantation of C. multijuga is being developed in the Ducke Forest near Manaus to try and understand the mechanism of the diesel oil formation in the trees, with the possibility of perhaps increasing the yield of this material. Also, it is hoped to learn whether or not it is possible to use more than one tap in each tree simultaneously to produce more oil. Because these trees take at least ten years to reach tappability, it will not be possible to use this candidate as a short-term choice; however, in the longer term in suitable areas of the world the Copaifera may turn out to be one of the best sources of oil from plants/trees that is available.

Isoprenoids from Plants and Trees

The oils from Euphorbias and Asclepias consist of a chemical similar to turpentine, being terpenes. Terpenes have the desirable characteristics which make the plants which produce them good energy agriculture candidates. For example, the black oil obtained from Euphorbia lathyris has been submitted to the shape selective zeolite catalyst developed by the Mobil Oil Corporation and has been found to produce ethylene, propylene, toluene, xylenes, nonaromatics, coke, alkanes and fuel oil, all useful for petrochemical industrial processes.¹²

The oil from the C. multijuga has been analyzed and is also a terpene, again directly useful in automobile engines as well as being a component of pharmaceuticals and used for medicinal purposes by the natives of the Amazon. Another type of oil is produced by another Brazilian tree, Marmeleiro (Croton, a member of the Euphorbiaceae) and terpenes are obtained from that as well, by steam distillation.

When we were in the Philippines recently we learned of a tree there which produces seeds containing oil. The fruit of this particular tree, Pittosporum resiniferum, is quite large and is used as a source of illumination by tying it to the end of a stick and lighting it.¹³ We obtained some P. resiniferum seeds and analyzed the "oil" from the fruits of these seeds, called petroleum nuts. This seed oil also contains terpenes and is another possible candidate for fuel and materials.¹⁴

Having analyzed the oil from P. resiniferum, we recognized that the same genus grew in California, but a different species. We performed some experiments with our own native, Pittosporum undulatum. The fruits of the California plant are much smaller and the seeds are very tiny inside the pod. The whole fruit is harvested, extracted with a solvent and the hexane extract contains terpenes similar to those from the P. resiniferum in the Philippines.

Conclusion

The notion of using plants as alternate energy sources is not new.^{15, 16} The idea of extracting oil from green plants and seeds is to find materials that would be hydrocarbon-like in nature, as opposed to the seed oils which are glycerides. The point also is to be able to use land that is not suitable for food production, and many of the Euphorbias are capable of growing in semiarid land with a minimum amount of water.

Two earlier attempts to use this idea were made in Africa, one by the French in Morocco in about 1940 where they used Euphorbia resinifera to produce latex from which were extracted gum resin and rubber.^{5c, d} Also, in Ethiopia the Italians got the idea that they might be able to harvest Euphorbia Abyssinica (Fig. 14), which grows prolifically in Ethiopia, to produce fuel oil and eventually gasoline.^{5d} However, they did not complete the project, but the idea itself was certainly feasible. In the one reference to this experiment that we have been able to find¹⁷ the statement is made that "the experiment will be watched with interest since geologists inform us that the world's

petroleum supply is limited and diminishing rapidly, and who knows but that the answer rests with the Euphorbiae." That statement is just as true today.

I think the time is now coming when we will again return to plant hydrocarbons as sources for chemicals, first, and eventually for fuels on a sustained basis. There are problems associated with this, of course, such as yield of products. Future work will involve not only classical methods of plant breeding and selection but also the newer techniques of genetic engineering to improve not only the plant itself but change the chemical composition of the components of the oil/chemicals that the plant produces. We will then be returning to using the green plant, a solar energy capturing device, to produce hydrocarbons of suitable molecular weight and structure which can be used for both materials and liquid fuels.

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Figure Captions

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U.S. ENERGY SOURCES AND USES IN 1980 (IN QUADRILLIONS OF BTUs)

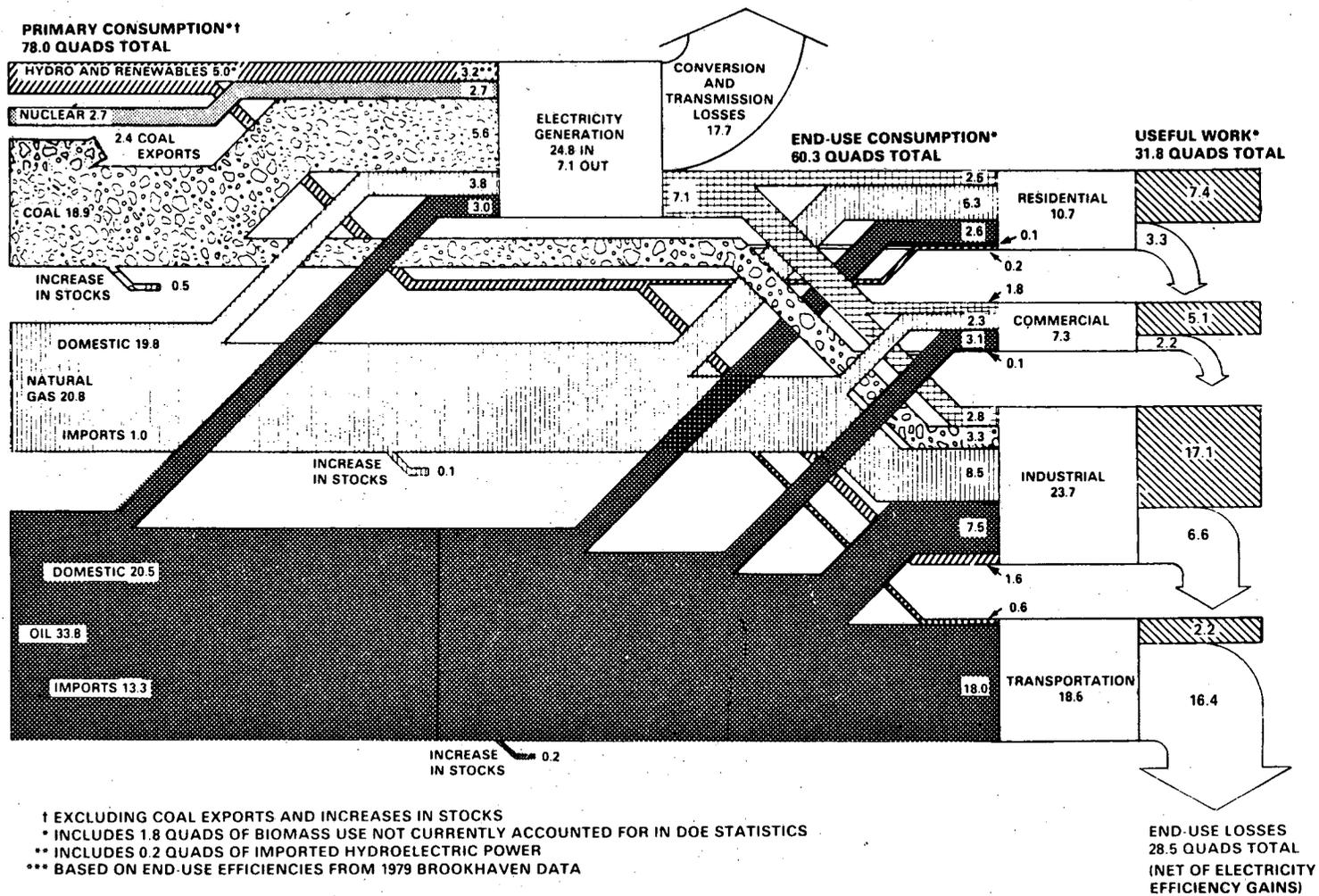
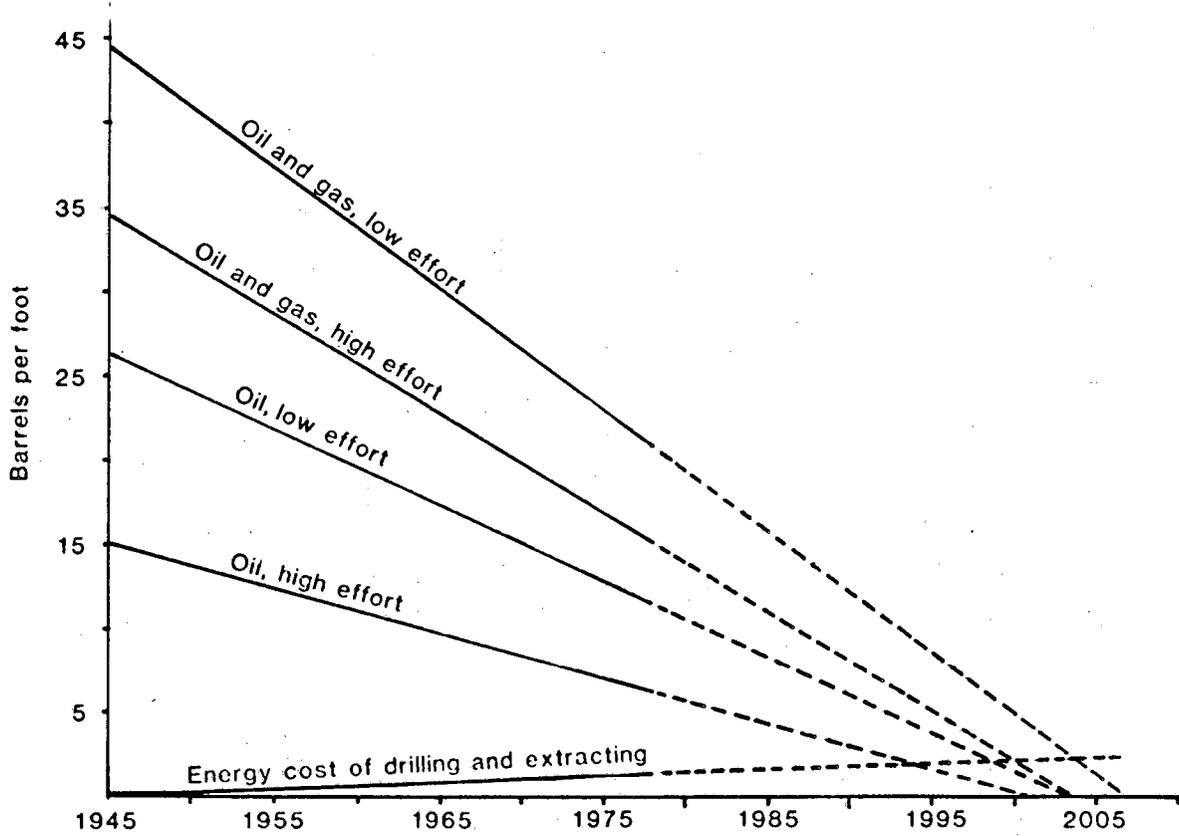


Fig. 1

XBL 818-11422



Linear extrapolations (dashed lines) of energy gains and energy costs for high and low drilling intensity. (Hall, 1981)

Fig. 2

XBL 813-8448

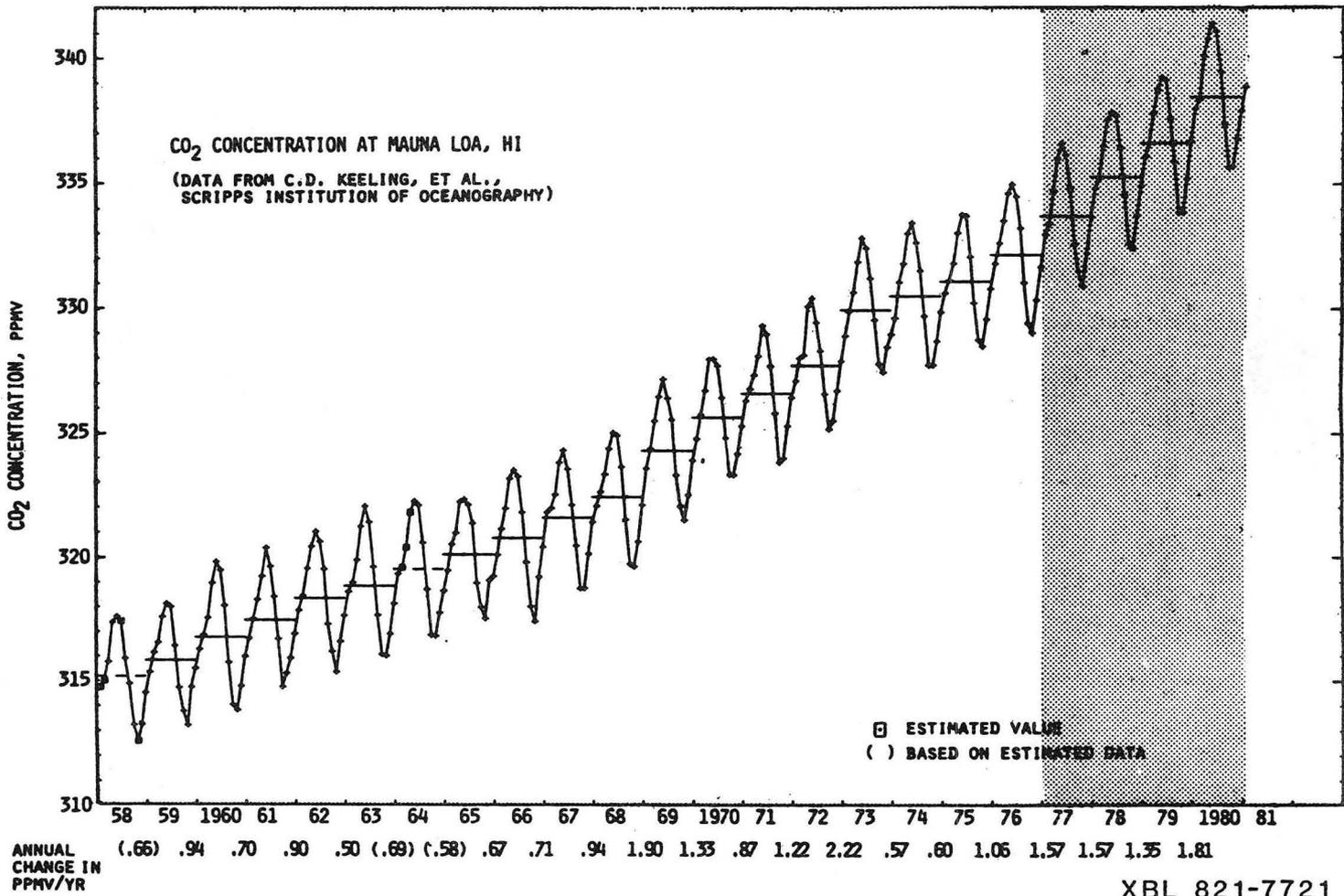


Fig. 3

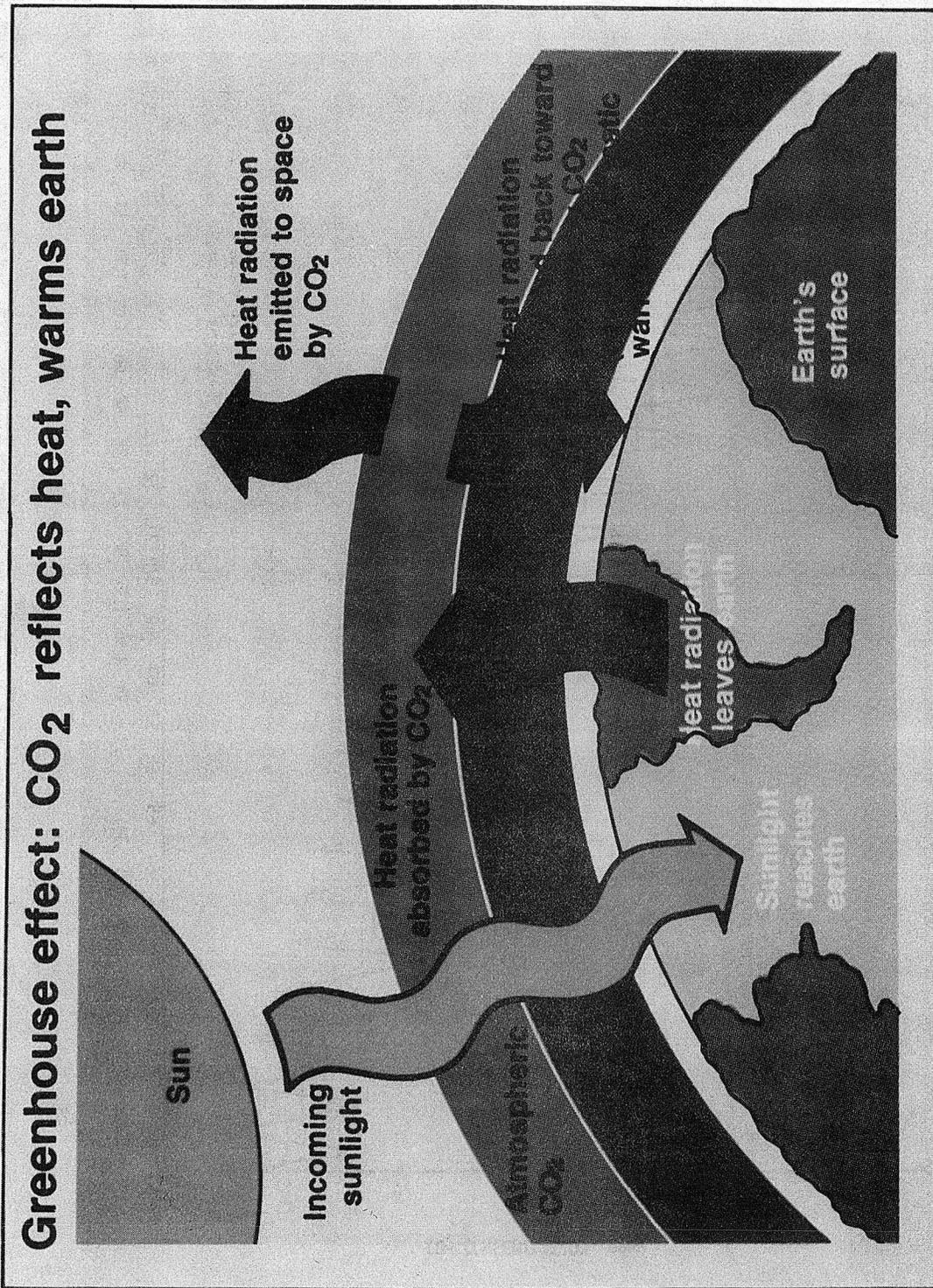
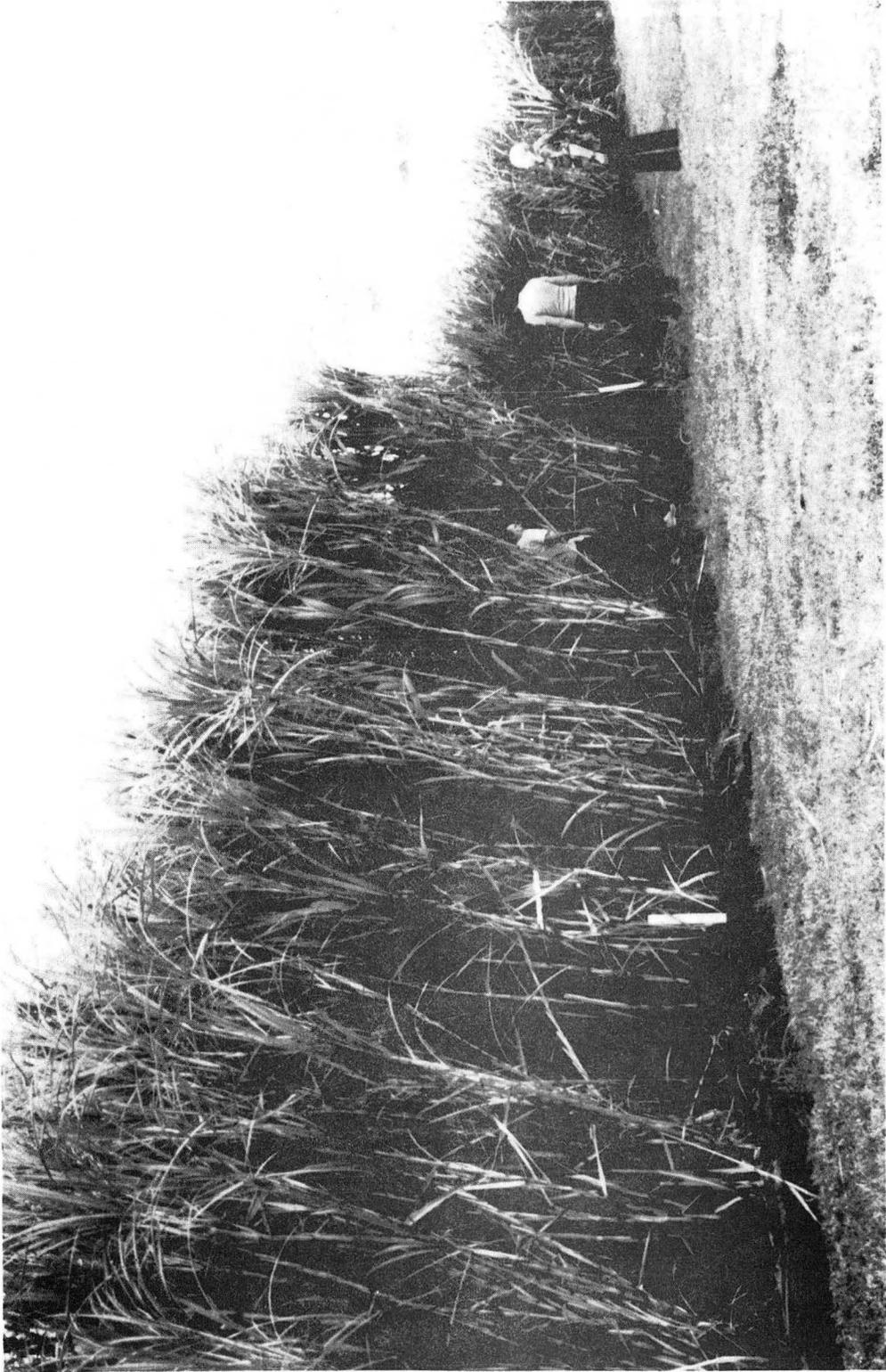


Fig. 4

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Fig. 5



Fig. 6

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Fig. 7

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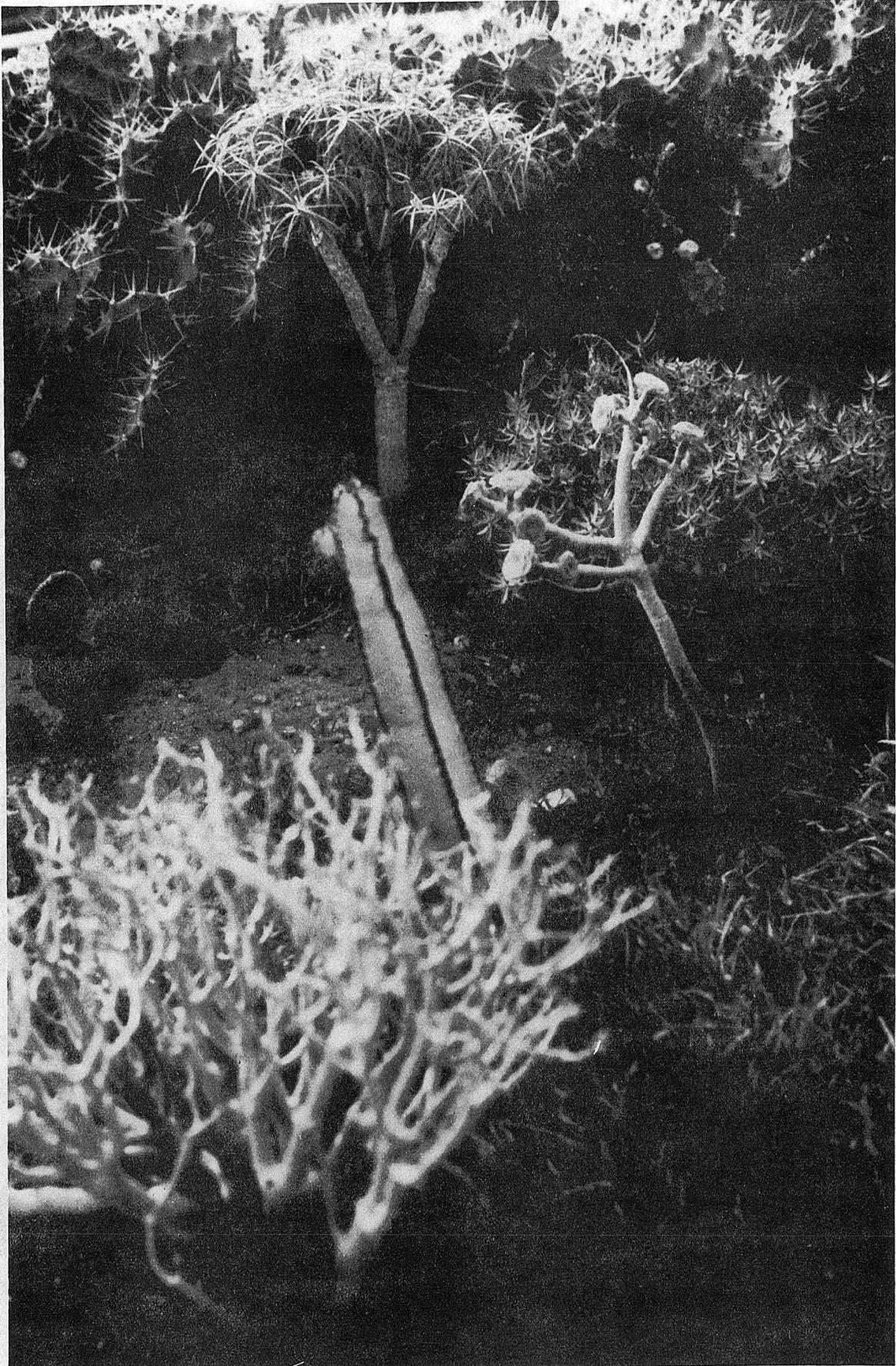


Fig. 8

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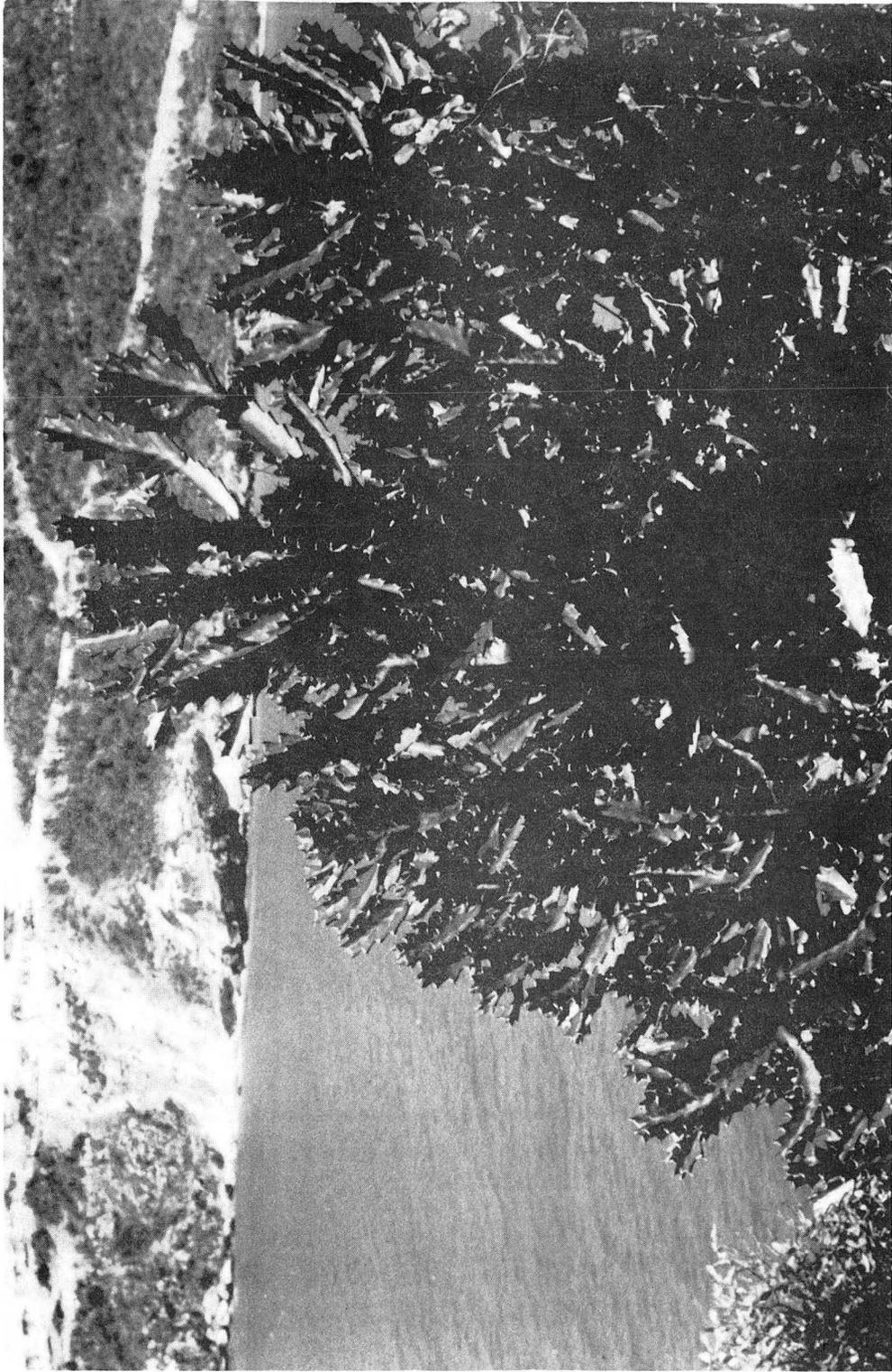
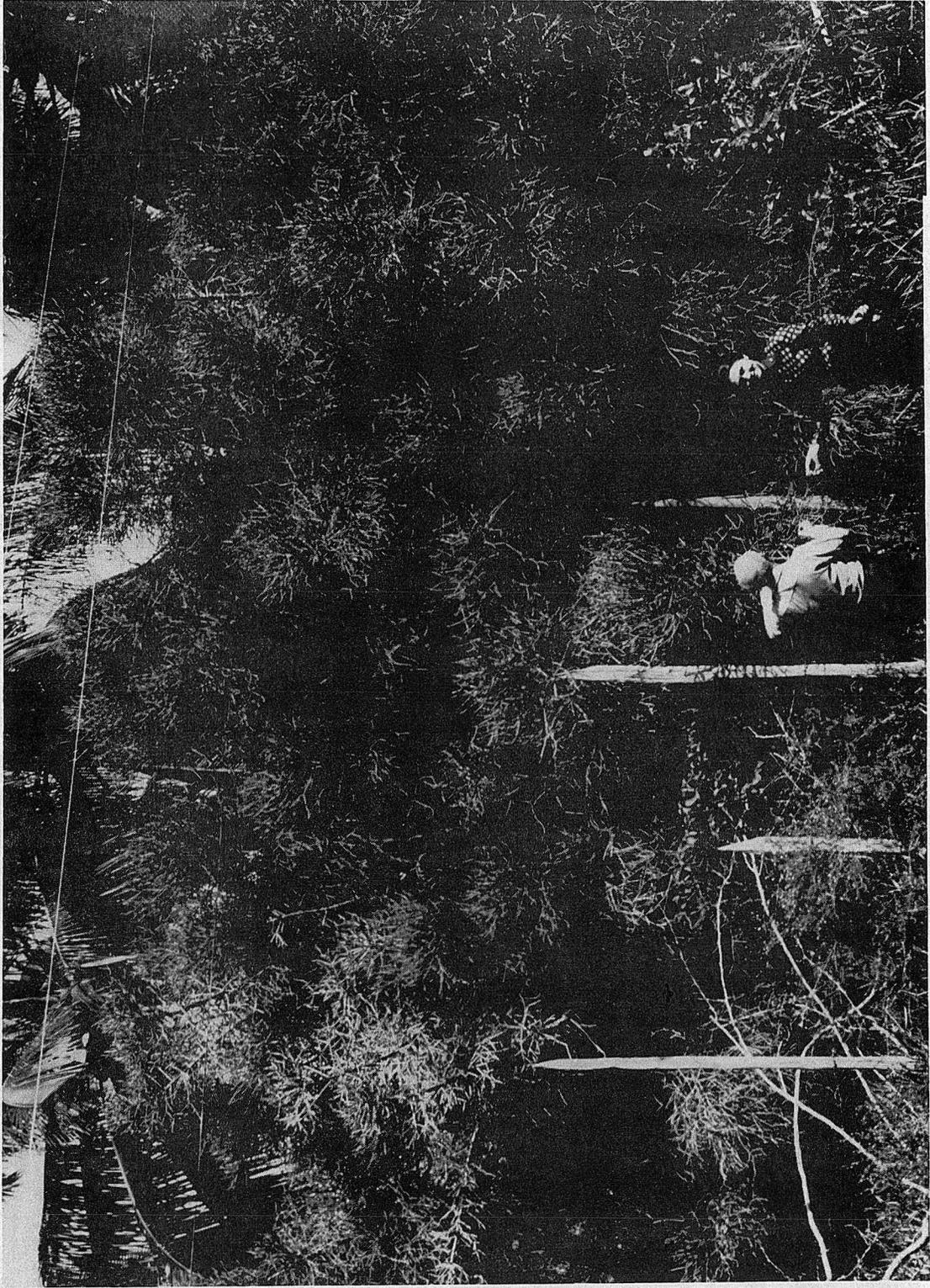


Fig. 9

BBC 775-5035



BBC 768-7678

Fig. 10

Conceptual Processing Sequence to Recover Terpenoids and Sugars from *Euphorbia lathyris*

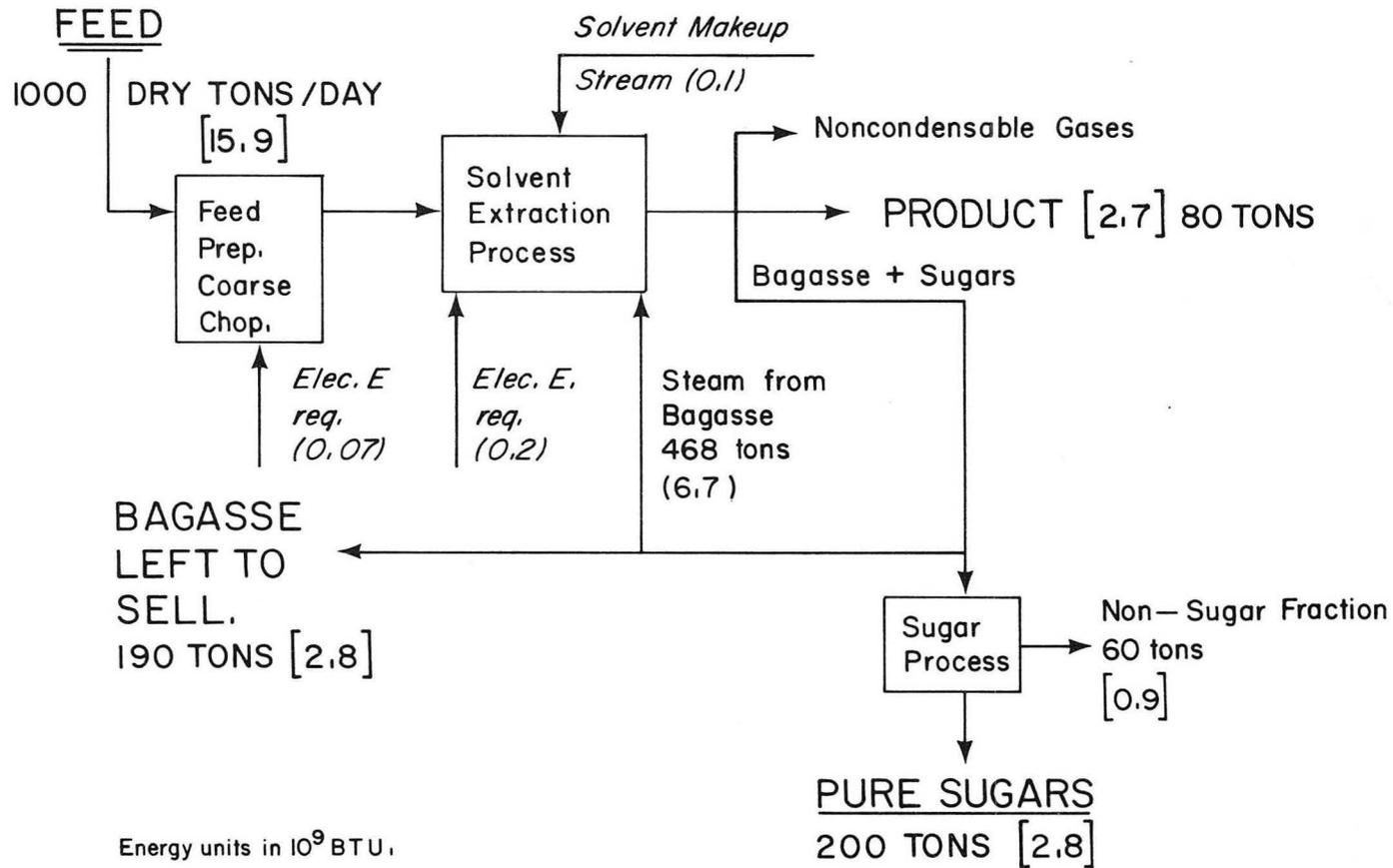


Fig. 11

XBL 807-4263A



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Fig. 12



Fig. 13

BBC 810-9901

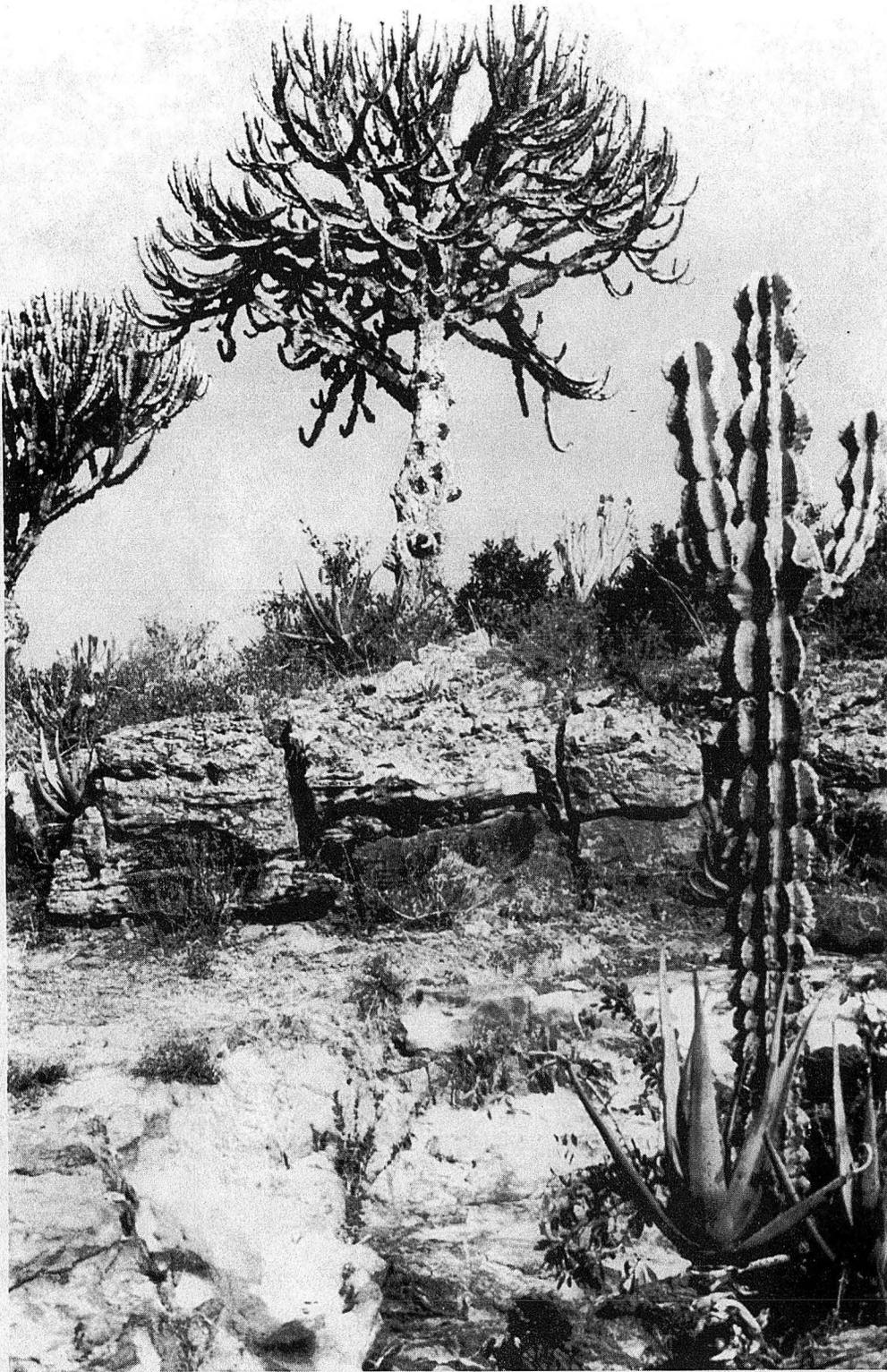


Fig. 14

CBB 792-2292

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