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ENVIRONMENTAL CONTROL TECHNOLOGY FOR ATMOSPHERIC CARBON DIOXIDE

QUARTERLY PROGRESS REPORT NO. 1
JUNE 20, 1977 TO SEPTEMBER 30, 1977

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TABLE OF CONTENTS

	<u>page</u>
ABSTRACT	iv
INTRODUCTION	1
RESULTS	
Literature Review	3
Impact of US Emissions	5
Impact of Coal Utilization	5
Process Considerations for CO ₂ Control	6
WORK FORECAST FOR NEXT QUARTER	8
REFERENCES	9
BACKGROUND LITERATURE	9
TABLE	10
FIGURES	11

ABSTRACT

The primary objective of the subject program is to assess the potential options for controlling atmospheric CO₂. Accordingly, CO₂ control scenarios based on conventional technology and applied to the larger industrial emitters will be prepared. The studies will include preliminary cost estimates of selected processes, to identify fruitful areas for environmental control technology (ECT) programmatic development as related to CO₂ release and control. BNL's prior experience in this area includes the development and evaluation of a number of processes for removing CO₂ from the atmosphere for the purpose of producing synthetic carbonaceous fuels including methanol, gasoline, and methane. Background information from other DOE programs will be used to determine the limitations for the control studies.

This progress report presents background information on: (1) the concentration levels of CO₂ in the atmosphere during the last one hundred years; (2) the possible effects of rising CO₂ levels; (3) the impact of fossil fuel use in the United States on overall worldwide CO₂ emissions; (4) the impact of increased coal utilization on CO₂ emissions; and (5) process considerations for controlling CO₂.

INTRODUCTION

The concentration of atmospheric CO₂ has steadily increased from about 295 ppm by volume in 1860 (pre-industrial revolution value) to the current value of 331 ppm. It is projected that the concentration of CO₂ in the atmosphere could reach several times its present value within the next one hundred years. Estimates of the anticipated biogeochemical effects of this increase range all the way from acceptable to catastrophic. Of primary concern is the warming ("greenhouse") effect which could be produced. While CO₂ is transparent to incoming solar radiation, it absorbs a portion of the infrared radiation returning to space from the earth. The amount of warming produced by a given CO₂ concentration increase is still uncertain and difficult to predict at present since it depends on many factors. Increased concentrations of particulate matter (and aerosols) in the global air mass could produce a cooling effect by backscatter of incoming solar radiation and thus counteract, to some degree, the warming produced by increasing CO₂ levels. However, environmental emission regulations on particulates, as well as on other identified pollutants, have become more and more stringent during recent years and the trend is expected to continue. Furthermore, to deliberately attempt to counteract the effects of CO₂ by allowing increased emissions of particulate pollutants does not appear to be a prudent strategy.

Currently, 5×10^9 metric tons of carbon per year are discharged to the atmosphere as CO₂ from worldwide sources. About 50% of the CO₂ produced by the combustion of fossil fuels can be accounted for by the concentration

increase of the atmosphere. It is assumed that the excess is stored in the oceans and in the terrestrial biomass although this balance is by no means certain. What portion is taken up by the oceans and what portion by the land biota is still undecided. Extensive studies currently being performed, and in planning stages, are being sponsored by the Environmental Office of the DOE to resolve these and other uncertainties concerning the carbon cycle and the possible climatic effects produced by CO_2 . However, there is a chance that naturally occurring and/or anthropogenic opposing factors will not just counter the effects of increasing CO_2 levels, and, therefore, it may be necessary to limit industrial CO_2 emissions in the future. Up to the present, there has been practically no concern about the health or environmental effects of CO_2 discharge; accordingly, there are no regulations concerning CO_2 emissions. Options for CO_2 control have received little attention and although preliminary evaluations indicate that control is theoretically possible, it may not be technically or economically practicable. It thus becomes important at this time to examine CO_2 control options to determine the technical feasibilities and the economic costs, should the need arise for CO_2 control. This forms the rationale for the subject program.

The objectives of the BNL study are: (1) to review background information on CO_2 emissions and atmospheric concentrations for the purpose of establishing boundary conditions for the control studies, (2) to review methods of recovery, reuse, and disposal of CO_2 contained in the atmosphere and in the effluents of energy related industrial streams, (3) to develop CO_2 control scenarios based on available technology, and (4) to identify fruitful areas for ECT programmatic development relating to CO_2 release and control.

RESULTS

Literature Review

The literature on past studies and current CO₂ programs have been reviewed to gain insight into and to assess the bounds of the potential CO₂ problem. Our findings are summarized below.

During the last one hundred years or so, the release of fossil carbon as CO₂ has increased at an exponential rate (Figure 1).¹ As a result, the concentration of CO₂ contained in the atmosphere has grown by about 12%, increasing from about 295 ppm by volume in 1860 (pre-industrial revolution) to the current level of 331 ppm. Except for short periods during World Wars I and II and the depression, the release rate of fossil carbon has increased at the rate of about 4.3% per year.² At present, with no CO₂ emission controls, about 5×10^9 metric tons of carbon per year as CO₂ are emitted to the atmosphere by the burning of fossil fuels from worldwide sources. About 50% of this carbon can be accounted for by the increase in the CO₂ concentration of the atmosphere. The remaining carbon is assumed to be absorbed by the oceans and by the land biota, in unknown proportions.² Deforestation considerations and the possibility of the release of CO₂ from the Pacific Ocean waters south of the equator due to the "Southern Oscillation",³ a naturally occurring phenomenon of unknown cause, tend to becloud the issue. A better understanding of the carbon cycle is definitely needed.

If the use of fossil fuels continued to grow at 4.3% per year until the estimated supplies thereof were exhausted, the use rate at the end of this period would be almost 64 times the current use rate, and the total CO₂ injected into the atmosphere (during this period) would be

about 12 times the pre-industrial content of the atmosphere.² Obviously, such a use pattern cannot be tolerated; however, based on more realistic assumptions, it is predicted that the concentration of atmospheric CO₂, relative to the pre-industrial value, could increase by a factor of from about 2 (low use case) to about 5 (high use case) during the next one hundred years.² The low use case corresponds to a fossil fuels growth rate of 2% per year until the year 2025, followed by a symmetrical decrease as alternative and renewable energy sources become more available and the use of fossil fuels is discouraged. The high use case corresponds to an initial annual growth rate of 4.3% with subsequent reductions in proportion to the ultimate fossil fuel supply that has been consumed. In both cases, about 50% of the emitted fossil carbon is assumed to be taken up by the oceans and land biota.

Of primary concern is the warming ("greenhouse") effect which could be produced by these increased CO₂ concentrations due to the absorption by CO₂ of a portion of the infrared radiation returning to space from the earth [the infrared absorption spectrum of CO₂ shows strong absorption bands at 667 cm⁻¹ (15 μ) and 2349 cm⁻¹ (4.26 μ)]. Additional effects caused by increased CO₂ levels could influence the circulation cells and precipitation regime (the regular pattern of seasonal rainfall) as well as the earth's albedo (reflection power).⁴ The amount of warming produced by a given CO₂ concentration increase is estimated at 1 to 5°K per doubling of the CO₂ concentration.² For the low use case described above, this corresponds to a mean global temperature rise of from about 1 to 5°K over the next one hundred years and from about 2 to 10°K for the high use case. A temperature rise of 1 or 2°K during this period may be acceptable,

however, a rise of 10°K would probably cause catastrophic effects.

Impact of US Emissions

In order to determine the impact of fossil fuel use in the United States on overall (worldwide) CO₂ emissions, we have superimposed several points on Figure 1, based on US fuel use data for the years 1950, 1955, 1960, 1965, 1970, 1973,⁵ and 1976,¹ to produce Figure 2. The points were obtained by subtracting the estimated emissions of carbon (as CO₂) by US sources from the corresponding worldwide emissions for each of the above years, and thus indicate the quantities of carbon that would have been emitted if the US had not burned any fossil fuels during these years. The reduction in carbon (and thus in CO₂) by this hypothetical non-use of fossil fuels in the US ranges from 0.32 ppm in 1950 to 0.64 ppm in 1973 (and in 1976). However, during this period the percentage of the total yearly emissions contributed by the US decreased from 43% in 1950 to 27% in 1976. Furthermore, it is projected that the contributions to global CO₂ production by the US and Canada will decrease to about 8% by the year 2025. Developing nations will account for 36% of the CO₂ production, the USSR and E. Europe 27%, Asia 19%, W. Europe 7%, and Japan and Australia 3%.¹ Therefore, it would appear that a worldwide effort would be required to effectively control the level of atmospheric CO₂. A unilateral effort by any one nation probably would not be very effective.

Impact of Coal Utilization

Table 1 contains a breakdown of the carbon (CO₂) produced from major worldwide sources during the years 1970 through 1976.¹ For the year 1976, using the calculated values of 18,000 BTU/lb of contained carbon

for coal, 22,400 BTU/lb carbon for fuel oil and 31,500 BTU/lb carbon for natural gas, and assuming that all fossil energy was produced by the burning of solid coal, the carbon release to the atmosphere would have been roughly 6×10^9 metric tons or about 1.2 times the quantity actually emitted. Energy differences required in mining versus drilling, preparation, transportation, etc. and in combustion efficiencies were not taken into account. Note that the weight ratio of CO_2 produced by the burning of coal to that produced by the burning of natural gas, for the same energy output, is about 1.8 and for coal versus fuel oil about 1.2. If, in the above calculation, we had assumed that the natural gas and petroleum consumed in 1976 had been replaced by coal derived gaseous and liquid fuels, the quantity of carbon released to the atmosphere would have been even greater than 6×10^9 metric tons, due to inefficiencies in the gasification and liquefaction processes. Thus, added reliance on coal for energy production could compound the atmospheric release rate of CO_2 , which has been increasing exponentially since 1860.

CO_2 Process Considerations for Control of CO_2

Based on the foregoing, there is a possibility that CO_2 control, at least to some degree, will be necessary in the future. The three most important factors, which will influence the technology and economics of any CO_2 control system are: (1) the quantity of CO_2 to be removed, (2) the concentration of CO_2 in the carrier stream(s), and (3) reuse/disposal alternatives of captured CO_2 . The first two parameters determine the quantities of gases and liquids to be handled, thus the size of the operation and the number of required sites, and the choices of removal processes.^{6,7}

Furthermore, the removal process can have a profound effect on the operation of the CO_2 producing process, itself. Reuse/disposal feasibilities of captured CO_2 are also of paramount importance. Some options are to recycle CO_2 into useful products such as carbonaceous fuel using a non-fossil energy source. Another is to dispose of captured CO_2 either terrestrially or extraterrestrially, if possible.

A practical example of a gaseous pollution problem presently being dealt with is the removal of SO_2 and particulates from power generating facilities. For this case, the cost of the pollution abatement equipment amounts to about 20% of the capital cost of the plant. Personnel requirements, plant operating costs, site size, and the constraints on site selection are also increased. Furthermore, disposal of the calcium sulfate and sulfite sludges produced in the SO_2 scrubbers presents a formidable environmental problem in itself.

With regard to CO_2 , three potential control points (sites) are available for removing it from the environment: (1) the stacks of fossil power and industrial plants; (2) the atmosphere; and (3) the oceans. The first has the advantage of dealing with relatively high concentrations of CO_2 , while the latter two provide more flexibility for site selection and simpler disposal (or recovery and reuse) logistics.

The choice of control point, as with the three factors listed above, will reflect on the size of operation, the number of required sites, the removal efficiency, the thermal requirements, the comparative economics and on the overall feasibility of operation. All of these factors, therefore, must be carefully examined and weighed in formulating CO_2 control scenarios. These will be reported

on during the next period of this study.

WORK FORECAST FOR NEXT QUARTER

1. Develop CO₂ control scenarios.
2. Compile input (performance and engineering data) on CO₂ control processes for utilization in the scenarios presented.

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Table 1

CALCULATED WORLD-WIDE CO₂ EMISSIONS

(Reference 1)

	<u>1970</u>	<u>1971</u>	<u>1972</u>	<u>1973</u>	<u>1974</u>	<u>1975</u>	<u>1976</u>
Coal	2160	2134	2163	2203	2237	2347	2418
CO ₂ Produced	1497	1479	1499	1527	1550	1626	1676
Lignite	855	864	876	886	887	898	940
CO ₂ Produced	237	239	243	246	246	249	260
Crude Petroleum	2270	2402	2532	2777	2791	2650	2801
N.G. Liquids	74	77	83	88	87	90	91
CO ₂ Produced	1803	1907	2011	2203	2213	2107	2224
Natural Gas	1040	1113	1172	1233	1257	1269	1288
CO ₂ Produced	545	583	614	646	659	665	675
Natural Gas Flared	167	173	186	204	209	198	210
CO ₂ Produced	90	94	100	110	113	105	113
Cement	598	639	678	690	692	728	723
CO ₂ Produced	82	88	93	95	95	100	99
Total CO ₂ (carbon)	4253	4389	4559	4826	4876	4852	5047
(ppm)	2.00	2.07	2.15	2.27	2.29	2.28	2.38

(All entries in 10⁶ metric tons except Natural Gas and Natural Gas Flared in 10⁹ cubic meters.)

Figure 1. CO₂ production from fossil fuels and cement (Reference 1).

Figure 2. CO₂ production from fossil fuels and cement. Solid lines - world production; Broken curve - world production less emissions by U.S. sources.

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