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K/EM-469

SANITIZED VERSION OF PROPOSED AIR POLLUTION PROGRAM REPORT DATED 5/28/57

(Sanitized Version of Unnumbered CRD Document)

Compiled by
S. G. Thornton
Environmental Management Division
OAK RIDGE K-25 SITE
for the Health Studies Agreement

July 22, 1996

Oak Ridge K-25 Site
Oak Ridge, Tennessee 37831-7314
managed by
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
for the U.S. DEPARTMENT OF ENERGY
under Contract DE-AC05-84OR21400

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This document has been approved for release to the public by:

for W. S. Ewart 8/8/96

Technical Information Officer Date
Oak Ridge K-25 Site

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INTER-COMPANY CORRESPONDENCE

UNION CARBIDE NUCLEAR COMPANY

A Division of Union Carbide and Carbon Corporation

To: Mr. L. B. Emlet
Building K-1001

Plant: ORGDP

Date: June 3, 1957

Subject: Air Pollution Program

Copies To: Mr. K. W. Bahler
Mr. R. G. Jordan
✓Mr. D. M. Lang
Mr. R. W. Levin
File - NoRC

The attached interim report, prepared by Dr. J. S. Lyon and our Industrial Hygiene staff, is supplied with the thought of keeping you abreast of our efforts in developing an appropriate air pollution program for the gaseous diffusion plants. Lack of universally acceptable standards, methods, and understandings of the effects of fluorine and fluorides, particularly with respect to vegetation damage, has retarded our progress in this endeavor. Further, this report does not include certain data currently being collected which may have a significant effect on the program.

As you undoubtedly remember, the initial problem or assignment was to devise a sound control program for monitoring the fluorine concentrations in and around the gaseous diffusion plants from the standpoint of damage to humans, livestock, or vegetation, as a result of visible damage to vegetation near the plant area. This was later expanded to include all air contaminants of toxicological significance. It was also agreed that, in order to simplify the problem, early efforts would be devoted toward a consistent program for the Paducah and Oak Ridge gaseous diffusion plants, and that this might later be extended to include the Portsmouth facility.

An appropriate air control program of this type is considered desirable for a number of reasons. It is needed to put the Company and the Commission in a position to assure protection of employees' health, to provide an objective defense against false allegations of harm or damage from toxic air contaminants, and to avoid a psychological situation wherein employees see dead vegetation at and near the plants and hence become unduly concerned about their own health.

While it will undoubtedly be necessary to continue our current program of urinary sampling and air sampling at work locations and other points considered to be of significance as an essential part of our health protection program, a sound overall air pollution monitoring program appears desirable to clearly establish an environmental control program independent of clinical data.

It is recognized that our progress in this endeavor has not fulfilled our hopes or desires. However, real progress has been made in several

specific areas. Some examples are:

1. It has been clearly established that, to the best of our knowledge, employees are not being harmed by fluorine.

2. We have compiled good estimates of amounts of significant plant effluents and the sources from which they are being released.

3. There has been a major reduction in the amount of fluorine released at ORGDP and Paducah. (See page 10 of the attached report)

4. Our personnel working on the program have come to a better understanding of the problems associated with the release of fluorine to the atmosphere and of the production and research efforts and results of other companies and institutions that are working in this relatively new field.

I plan to make the necessary arrangements to assure that we vigorously pursue the task of getting agreement of Medical, Industrial Hygiene, and Laboratory personnel on (1) mutually acceptable, standard sampling and analytical methods, and (2) a more specific sampling schedule to be consistently followed at Oak Ridge and Paducah, which will provide appropriate in-plant and out-of-plant coverage to fulfill legal and psychological needs.

for A. P. Huber

APH:KWB:ngwk

Enclosure

~~BUSINESS - CONFIDENTIAL~~

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1957 JUN -7 AM 9:13

PROPOSED AIR POLLUTION PROGRAM

Interim Report

May 28, 1957

UNION CARBIDE NUCLEAR COMPANY
UNION CARBIDE AND CARBON CORPORATION
Oak Ridge Gaseous Diffusion Plant
Oak Ridge, Tennessee

Union Carbide Nuclear Company, Oak Ridge Gaseous Diffusion Plant, Operating Contractor for the U.S. Atomic Energy Commission.

May [illegible], 1957

INTRODUCTION

The possibility of air pollution from ORGDP effluents in concentrations toxic to vegetation became of more pronounced general interest following the observation in late 1955 of extensive marking of foliage east of the K-1037 Building. Prior to this time, air pollution work as such, had at both Paducah and ORGDP, been confined to specific acute problems and the great bulk of the organized air-survey program had been directed toward measuring air contaminants in work locations. Following this observation, rather extensive surveys by TVA foresters,[1] outlined further areas of vegetation marking and some areas of actual damage. It was their impression that, with the exception of the trees east of K-1037, most of the actual damage was due to deliberately used exfoliants along power lines and rights of way, with some damage due to a tip moth infestation. The markings, they felt, were probably from plant effluents with hydrogen fluoride considered to be the most likely offender. The damage east of K-1037, in view of the later regeneration of foliage, was attributed to an acute, massive release rather than a chronic state of air contamination. Similar observations were made at the Paducah plant where the problem was given greater urgency by their location in the midst of privately owned land, including grazing land, and by the greater magnitude of their fluoride effluents.

THE PROBLEM

It was decided at both plants that, since air pollution studies on a routine basis were indicated, they should be broadened to include all plant effluents of toxicological significance. Accordingly, after multiple conferences, visits to and by "experts" and extensive literature searching, the two plants reached an agreement on the problem involved and the approaches to be used.

The problem was felt to have two main components: (1) the possibility of employee injury and (2) vegetation damage and its concomitant problem of injury to grazing animals. The first component has already been settled. In the opinion of the two medical directors, voluminous negative urinary data have been established at both plants that employees are not being exposed to contaminants in concentrations of any human toxicological significance. This is further substantiated by some in-plant environmental surveys of fluorine concentrations in the atmosphere.[6] The vegetation component is a complex one and an approach to it made it desirable, in view of the differences in opinion of "experts" and the acknowledged ignorance of the effects of fluorine itself on vegetation, to make two assumptions: (1) that the Boyce Thompson Institute[7, 8] opinions on fluoride vegetation pathology are the most accurate and their investigative techniques and equipment the most reliable and (2) that fluoride levels in which fluorine is one component have the same significance as fluoride levels which do not contain fluorine.[9] If this latter assumption is correct, and neither plant believes it to be entirely true, it would follow from Boyce Thompson studies that findings of above 2 ppb of fluoride in air indicate a hazard to sensitive vegetation, while concentrations below this are probably safe.[9]

PROPOSED PROGRAM

An exploratory type of monitoring program will be utilized, initially at least, as the most economical method of evaluating the over-all air pollution problem. It will be designed primarily to detect atmospheric fluorides, sulfur dioxide, and boron as described in the following paragraphs. The influence of other possible pollutants, chlorine and hydrocarbon combustion products, will

be determined as a secondary part of the program. For legal purposes only, a composite of those samples taken for fluorides will be analyzed for uranium.

Two samplers will be employed near the perimeter fence for the collection of atmospheric fluorides. They will collect shift length samples on a continuous basis and their positions will be changed once a day. Moving around the plant, they will complete a circuit of the plant in approximately 8 days. Additional samples may be taken where sample results indicate the need. Paducah will also utilize an exploratory pattern of air sampling plus vegetation sampling of surrounding private land.

Sulfur dioxide in the atmosphere will be collected in an automatic impinger, already available in the works Laboratory,[16] in one location only - near the guard tower, K-1037-S-5, southeast of K-1400 initially. This is the most likely site for heavy concentrations of sulfur dioxide and is near the point of maximum vegetative marking which might be attributed to it. Paducah will await GDP developmental work on sulfur dioxide analysis before entering this phase of the program.

Soil samples for boron will be repeated east of K-1037 after a few weeks of dry weather have eliminated the leaching out problem. Boron is unique to the GDP melting operation.

The samples taken for fluorides will be checked for their chloride content, and, if significant, a more specific method for atmospheric chlorides will be developed.

Although the basic approach will be the same by both plants, there will be certain variations brought about by differences such as proximity to privately owned land, types and quantities of plant effluents, and the need for research and development in sampling, analytical and evaluation techniques. Paducah, for instance, because of its location, finds it necessary now to do vegetation sampling on grazing land, while ORGDP can afford the time for air surveys to determine the need for such sampling. To prevent duplication, certain development problems have been divided between the plants, with Paducah seeking to establish a correlation between air concentrations of fluoride effluent to plant damage and ORGDP trying to find a method of accurately analyzing sulfur dioxide concentrations in the presence of interfering agents.

It is planned that if air sampling results at ORGDP indicate a possible hazard to private farm land, the sampling program will be extended. Likewise, if preliminary studies of fluoride fall-out in ORGDP parking lots indicate a potential auto paint effect, further work may be desirable.

THRESHOLD LIMITS

Data will be interpreted in relation to the most authoritative threshold limits obtainable, since nationally recognized limits have not yet been established for vegetation toxicology.[22,27,28] Boyce Thompson values will be recognized for fluorides. Threshold limits that will be initially used for this study follow:

Sulfur Dioxide

TVA accepts the threshold value of 0.5 ppm sulfur dioxide for 30 minutes. That is, a 30-minute exposure at a level of 0.5 ppm causes marking on sensitive plants.

Chlorine

Injury occurs to beans and radish in 0.5 hour at 1.3 ppm, to roses in 0.5 hour at 1.5 ppm, to buckwheat in one hour at 0.46 ppm, and to peach in 3 hours at 0.26 ppm.[28]

Hydrogen Fluoride

Pine (ponderosa) young needles slightly injured after [illegible] 9 days fumigation with 5 ppb or less. Boyce Thompson Institute and State College, Washington, agree on this.[28] Pine (ponderosa, old needles) required more than 10 ppb as reported by State College, Washington.[28] White pine is in the same susceptibility class as old needles of ponderosa pine.

Boron

Excess of 0.5 - 5.0 ppm free boron[12] is toxic to most plants. Dr. W. L. Parks of the Agronomy Department, University of Tennessee, states that 40 to 50 pounds of boron/acre is recommended as fertilizer for alfalfa in the State of Tennessee. As high as 100 pounds/acre have been used without injurious effects.

Uranium

No vegetation toxicity known. Human MAC - 0.05 mg/cuM.

CURRENT STATUS

The following table presents some idea of the comparative effluent problem at the two plants:

COMPARISON OF AMOUNT OF PLANT EFFLUENTS AT THAT OF PADUCAH

Material	Amount ORGDP	Amount Paducah
Fluorides (Gaseous)		
*Sulfur Dioxide	947,255 pounds/month	From 444,000 pounds/month to 1,068,000 pounds/month
Boron	100 pounds/month	None

Table I in the appendix presents detailed information on ORGDP effluents insofar as these are known. Of these effluents, sulfur dioxide (possibly the local power house and steam plant, and possibly from the Kingston steam plant), fluorides, chlorine, and boron[26] are considered to be possibly significant from a vegetation marking standpoint. Uranium is considered to have only legal significance. Materials listed in the table but not covered in the proposed program are considered to be quantitatively or inherently of no toxicological significance.

Map I in the appendix shows the areas of vegetation marking and areas of damage near the end of the 1956 growing season. The trees east of K-1037, which precipitated this study, had almost completely recovered by the end of 1956 but are now beginning to show markings again.

One sampler based on the Boyce Thompson design has been constructed at ORGDP and is now being field tested.[15]

A turbidimetric method measuring sulfur dioxide concentrations is being developed to circumvent interference from oxidants and acid gases which have made the usual measuring techniques useless.[17]

*These figures do not take into consideration the sulfur dioxide from Shawnee and Kingston Steam Plants.

INTERIM REPORT
May 23, 1957

Sixteen soil samples taken from the area east of the [illegible] shelter have been analyzed for their boron contents. Despite recent heavy rains which may have leached considerable amounts of boron out, only 4 samples showed no boron; the others ranged in value from 0.2 ppm to 0.7 ppm.[12,13,14]

SUMMARY AND CONCLUSIONS

The most charitable statement that can be made about the present status of air pollution evaluation as a science is that it is in its infancy; in fact, it is still almost a fetus. Detection and analytical techniques for many contaminants are still in the developmental stage and threshold limits for a tremendous number of materials simply have not been established; where they have, they vary, depending on the duration of exposure and the type of vegetation being exposed. It seems in general, however, that threshold limits for plants are lower than for human exposure; certainly this is true for the fluorides. This vast field of ignorance does not denote a hopeless situation but it does point up the necessity for a careful, conservative approach and for correlating air concentrations of specific contaminants with visual observation of vegetation effects during the growing season.

With regard to specific problems, it is felt that sufficient urinary and clinical data exist at ORGDP and at Paducah to positively rule out any injury to plant personnel from air contamination. The picture on vegetation injury is, unfortunately, not as clean cut although some generalizations seem safe.

At ORGDP the situation is less serious both from the standpoint of plant location and quantity of plant effluents than at Paducah. Vegetation injury at ORGDP is believed, by presumably competent foresters,[1] to be minimal and to be properly classified as vegetation marking rather than actual damage.

It is believed that if ORGDP is damaging vegetation, the offending material will be found to be either fluorides, sulfur dioxide, boron, chlorides, or a combination of these. Common principles of approach to the problem have been reached with Paducah and agreement reached that a limited, exploratory type of sampling, combined with visual observation of vegetation, will ultimately provide an answer to the vegetation problem, though not in the immediate future.

There is some evidence, not conclusive at all, that there may be a fluoride corrosion problem with automobiles in ORGDP parking lots. This probably deserves further investigation.

[1] See appendix for further details.

APPENDIX A

Sampling and Analytical Methods

Analytical methods now in common use at ORGDP and the Paducah plant will be used as much as possible. Where differences between the analytical methods used at the two plants do exist, these will be carefully scrutinized and, if necessary reconciled. Where special methods are necessary and no existing ASTM, etc., methods are available, some will be developed to fit the needs of both plants.

Air pollution samples for fluorides will be collected in the Boyce Thompson Institute impinger[7] of 500 ml capacity in 200 ml of a 0.5 per cent solution of sodium hydroxide in distilled water. A small aliquot of each 8-hour sample will be withdrawn before the sample is processed for fluorides. The fluorides will be steam distilled from the collectors in the manner described by the Boyce Thompson Institute.[7] The aliquots from each of the three 8-hour samples per location will be composited and one uranium determination per sample location per 24 hours will be made.

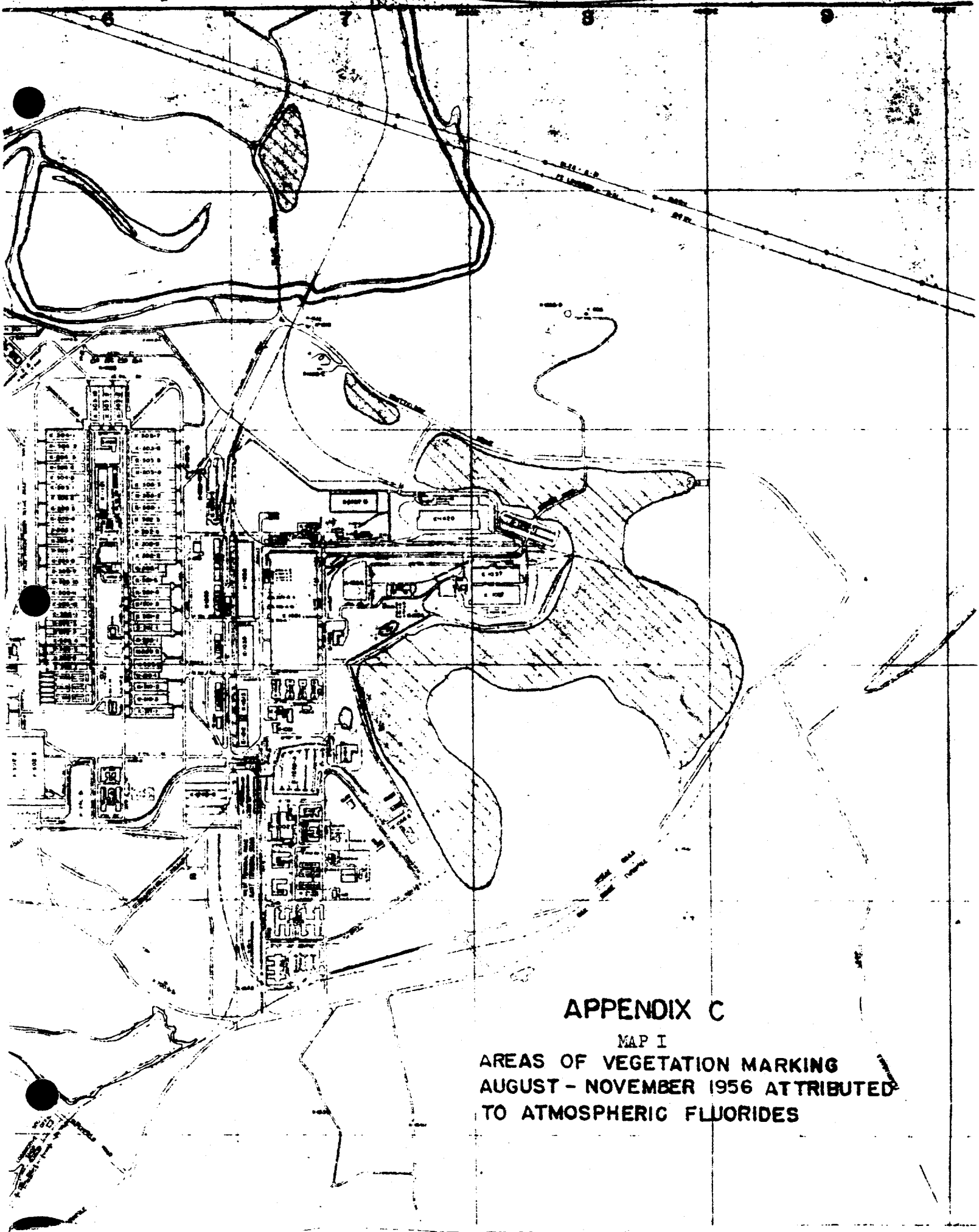
Costs will be difficult to evaluate until some field work is actually done and some intelligent evaluation of the magnitude of the problem can be made. It should be possible to start, however, with an expenditure of $2,000 for sampling equipment and $4,500 for labor.

~~BUSINESS - CONFIDENTIAL~~

May , 1957

History of Cascade Side Purge

The Cascade fluorine vent purge rate averaged 1,390 pounds/month from August, 1953, through September, 1953



APPENDIX C

MAP I

AREAS OF VEGETATION MARKING AUGUST - NOVEMBER 1956 ATTRIBUTED TO ATMOSPHERIC FLUORIDES

~~BUSINESS - CONFIDENTIAL~~ INTERIM REPORT
May 28, 1957

APPENDIX D

TABLE I

SURVEY OF ATMOSPHERIC EFFLUENT OF THE ORGDP - April 10, 1957

Building	Type Operation	Contaminant	Amount of Contaminant Emitted/Month Cal. as F_2 Except Where Indicated 1957	1954
K-31	Cold Trapping	F_2, HF, UF_6	102 lbs.	102 lbs.
K-33	Cold Trapping	F_2, HF, UF_6	516 lbs.	516 lbs.
K-33	K-33 Cooling Water	CrO_4	2,880 lbs.	Not using CrO_4
K-302-5	Purge & Product	UF_6, F_2	1 lb.	1 lb.
K-311-1*	Side Purge	F_2		
K-312-1	Side Purge	F_2		
K-312-3	Top Purge	F_2		
K-402-8	Cold Trapping	F_2, HF, UF_6	5.4 lbs.	11 lbs.
K-413	Cold Trapping & Side Withdrawals	F_2, HF, UF_6	5.4 lbs.	11 lbs.
K-631	Tails Withdrawal	UF_6	A release approx. once/yr.	A release approx. once/yr.
K-633	Test Loops	F_2	18 lbs.	18 lbs.
K-701	Power House	SO_2, F^-, Fly Ash	SO_2 released in lbs./mo. during 1957 are as follows: Jan., 5,016,200; Feb., 186,144; Mar., 27,806; Apr., 0.0; May, 0.0; June, 0.0; July, 0.0; Aug., 0.0; Sept., 0.0; Oct., 20,580; Nov., 2,771,160; Dec., 2,949,380.	SO_2 released in lbs./mo. during 1954 are as follows: Jan., 5,320,000; Feb., 2,980,000; Mar., 2,881,000; Apr., 653,000; May, 368,000; June, 42,000; July, 6,400; Aug., 0.0; Sept., 1,600; Oct., 1,110,400; Nov., 2,724,000; Dec., 4,105,200. These values will increase by 20% per year for 57, 58 and so.

~~BUSINESS - CONFIDENTIAL~~

INTERIM REPORT
May 28, 1957

SURVEY OF ATMOSPHERIC EFFLUENT OF THE GDP - April 10, 1957

Building	Type Operation	Contaminant	Amount of Contaminant Emitted/Month Cal. as F_2 Except Where Indicated 1957	1954
K-1004-L	UF_6 Pilot Plant	UF_6	Release of 1/2 lb. approx. 3 times yearly.	1/2 lb. 3 times/yr.
K-1004-L	UF_6 Pilot Plant	F_2	4.5 lbs./month	4.5 lbs./month
K-1004-L	UF_6 Pilot Plant	ClF_3	Trace quantities of ClF_3 released 3 times/mo.	Trace ClF_3 3 times/mo.
K-1037		Boron	100 lbs./mo.	100 lbs./mo.
K-1131*	Cold Trapping	UF_6	50 lbs. UF_6/mo.	62 lbs. UF_6/mo.
K-1131*	Feed Manufacturing	F_2, HF	100 lbs. F_2/mo.; 70 lbs. HF/mo.	250 lbs. F_2 and 1,250 lbs. HF discharged to stack/mo.
K-1131	Cleanup Reactor		407 lbs. F_2/mo.	Not in operation.
K-1302	F_2 Cells Conditioned	HF, F_2	3,620 lbs. HF/mo.	3,620 lbs. HF/mo.
K-1401	Converter Conditioning	F_2, ClF_3, UF_6	10 lbs. ClF_3/mo., 30 lbs. F_2/mo.	13 lbs. ClF_3/mo., 40 lbs. F_2/mo.
K-1410	Decontamination	UF_6	UF_6 releases possible on occasions.	UF_6 releases possible on occasions.

*There has been a reduction in effluent since 1954.

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[illegible] Report
May [illegible], 1957

SURVEY OF ATMOSPHERIC EFFLUENT OF [illegible] AREA - April [illegible], 1957

Building	Type Operation	Contaminant	Amount of Contaminant Emitted/Month Cal. as F_2 except Where Indicated 1957	1954
K-1413	LMD Pilot Plant	UF_6, F_2	20 lbs. UF_6/mo., 960 lbs. F_2/mo.	20 lbs. UF_6/mo., 960 lbs. F_2/mo.
K-1420	Nitric Acid Spray Booth	NO_2, NO, N_2O, N_2O_4	534 lbs./mo. as HNO_3	Nitric Acid Booth was operating in K-1303. 333 lbs./mo. as HNO_3.
K-1420	Furnace Stand Converter F_2 Conditioning	F_2	20 lbs./mo.	20 lbs./mo.
K-1420	Nickel Recovery	Ni	Trace possible.	Not in operation.
K-1420	Oxide Fluorination	F_2	159 lbs./mo.	Operating in K-1301 40 lbs./mo. Cal. as HNO_3.
K-1420	Drum Dryer Calciner	NO_2, NO, N_2O, N_2O_4	150 lbs./mo. as HNO_3	Not in operation.
K-1421	Incineration	Fly Ash	Possible Traces of UO_2F_2 and UF_4.	Operating in K-1410, possible traces of UO_2, F_2, and UF_4.
K-1501	Steam Plant	SO_2, F^-, Fly Ash	SO_2 released in lbs./mo. during 1956 are as follows: Jan., 152,460; Feb., 47,550; Mar., 34,181; Apr., 5,964; May, 979; June, 702; July, 0.0; Aug., 0.0; Sept., 14,147; Oct., 21,337; Nov., 54,156; Dec., 24,319.	SO_2 released in lbs./mo. during 1954 are as follows: Jan., 235,600; Feb., 59,200; Mar., 191,200; Apr., 84,000; May, 60,000; June, 800; July, 0.0; Aug., 0.0; Sept., 0.0; Oct., 0.0; Nov., 133,600; Dec., 332,400.

APPENDIX E

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