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NO_x EMISSIONS FROM HANFORD
NUCLEAR FUELS REPROCESSING PLANTS

A. L. Pajunen
Separations Process Support Unit
Separations Process Engineering Group
Process Engineering Department
Research and Engineering Division

R. L. Dirkes
Environmental Protection Group
Environmental and Occupational Safety Department
Health, Safety and Environmental Division

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APPROVED:

Wayne Ogren 9-14-78
Manager, Separations Process Support Unit

APPROVED:

Robert Paresch
Manager, Environmental Protection Group

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Rockwell International
Rockwell Hanford Operations
Energy Systems Group
Richland, Washington 99352

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NO_x EMISSIONS FROM HANFORD
NUCLEAR FUELS REPROCESSING PLANTS

EXECUTIVE SUMMARY

Operation of the existing Hanford nuclear fuel reprocessing facilities will increase the release of nitrogen oxides (NO_x) to the atmosphere over present emission rates. Stack emissions from two reprocessing facilities, one waste storage facility and two coal burning power plants will contain increased concentrations of NO_x. The opacity of the reprocessing facilities' emissions is predicted to periodically exceed the State and local opacity limit of twenty percent.

Past measurements failed to detect differences in the ambient air NO_x concentration with and without reprocessing plant operations. Since the facilities are not presently operating, increases in the non-occupational ambient air NO_x concentration were predicted from theoretical diffusion models. Based on the calculations, the annual average ambient air NO_x concentration will increase from the present level of less than 0.004 ppm to less than 0.006 ppm at the Hanford site boundaries. The national standard for the annual mean ambient air NO₂ concentration is 0.05 ppm. Therefore, the non-occupational ambient air NO_x concentration will not be increased to significant levels by reprocessing operations in the Hanford 200 Areas.

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NO_x EMISSIONS FROM HANFORD NUCLEAR FUELS REPROCESSING PLANTS

INTRODUCTION

Rockwell Hanford Operations, under contract to the Department of Energy, is preparing to start-up the Hanford Facilities for reprocessing spent nuclear fuels. Operation of the existing facilities will increase the release of nitrogen oxides (NO_x) to the atmosphere over present emission rates. Nitrogen oxide emissions from the facilities will be subject to Federal, State and local emission standards. Observations by plant personnel during previous operating periods indicate that some stack emissions will not comply with the twenty percent opacity limit. Therefore, a variance from the opacity emission standard will be required to allow operation of the existing facilities without additional NO_x abatement equipment.

Preliminary estimates indicate that the occupational ambient air NO_x concentration will remain within Federal guidelines, but, plant emissions may be subject to the Prevention of Significant Deterioration (PSD) regulations of the 1977 Clean Air Act Amendments which became effective after fuel reprocessing operations at Hanford were discontinued. The following report describes the NO_x sources related to fuel reprocessing in the Hanford 200 Areas and discusses the predicted impact of increased NO_x emissions on the ambient air quality of regions surrounding the reprocessing facilities. The discussion is intended to provide background information to support the application for a variance from the local opacity emission standard and aid in determining if plant emissions are subject to PSD regulations.

DESCRIPTION OF SOURCES

Reprocessing spent nuclear fuels at Hanford will result in an increase of NO_x emissions from five points or stacks in the 200 Areas. The source locations are shown on the map sketched in Figure 1. Emission points include two chemical processing plants, two coal-fired power plants and one waste storage facility. The source name, function, location and major constituent of the emitted NO_x are listed in Table I.

BACKGROUND INFORMATION

Nitrogen oxides are produced by the Hanford reprocessing facilities in the form of nitric oxide (NO) and/or nitrogen dioxide (NO₂). Nitric oxide is a colorless gas which reacts with oxygen to form nitrogen dioxide. Nitrogen dioxide is a brown colored gas which rapidly comes to equilibrium with the

FIGURE 1
Hanford Fuel Reprocessing NO_x Source Locations

colorless dimer, dinitrogen tetroxide (N_2O_4). The reaction sequence is summarized by

TABLE I

Hanford Fuel Reprocessing NO_x Sources

Name	Function	Location	Major NO_x Constituent
The Purex Plant (202-A Building)	Dissolves and processes spent fuel rods to separate fission products, uranium, neptunium and plutonium. The products are uranium and neptunium solutions and a plutonium oxide powder.	200-E Area	Nitrogen Dioxide (NO_2)
AR Vault (244-AR Building)	Accumulates treated acidic wastes containing fission products before further processing and storage.	200-E Area	NO_2
The UO_3 Plant (224-U Building)	Processes uranium solutions from the Purex Plant producing uranium oxide powder which is packaged for shipping.	200-W Area	NO_2
Power Plants (284-E and 284-W Buildings)	Burns coal to produce steam for process uses.	One in 200-E and one in 200-W Areas	Nitric Oxide (NO)

The opacity of an off-gas plume is dependent on: the concentration of color producing contaminant; stack diameter; angle relationship between the sun, plume and observer; background weather conditions; and the individual observer (1). Therefore, an increase in the concentration of the color producing contaminant (in the case of NO_x emissions the colored contaminant is NO_2) will increase the opacity of the stack off-gases. Table I shows that emissions from the Purex Plant, AR Vault and UO_3 Plant will be colored due to releases of NO_2 . The opacity of power plant emissions will not be affected by changes in the NO_x concentration since the major constituent of NO_x released is in the form of colorless NO . A quantitative NO_x concentration has not been associated with the twenty percent opacity limit for an off-gas plume. Therefore, the opacity of each plant plume is described in qualitative terms.

Regulations, concerning the Prevention of Significant Deterioration of air quality, were promulgated in 1977 to protect the national ambient air quality from significant incremental increases in pollutant content (2). The original regulations set limits for the maximum allowable increase of particulates and sulfur dioxide over baseline ambient air concentrations. At this time, no quantitative limit has been established for the impact of NO_x on the ambient air quality.

THE PUREX PLANT

The Plutonium Uranium Extraction (Purex) Plant separates uranium, neptunium, plutonium and waste fission products. Spent fuel rods are dissolved in nitric acid to form feed solutions which are processed, separating products from wastes in solvent extraction columns. A majority of the NO_x released from the Purex Plant is created by the dissolution of uranium metal and treatment of acidic wastes with sucrose. Smaller quantities of NO_x are evolved from the dissolution of recovered uranium dioxide (UO_2) and the conversion of plutonium solutions to plutonium oxide powder (plutonium denitration). Off-gases from the waste treatment, recovered UO_2 dissolution and plutonium denitration processes are combined with ventilation air exhausted from the main building. The building ventilation air is then combined with the dissolver off-gases and released via a stack, 7 feet in diameter and 200 feet high. A plot plan sketch of the Purex dissolver and main building ventilation system is shown in Figure 2.

Uranium Metal Dissolution

The initial processing or feed preparation steps of the Purex process convert batches of metal fuel rods into a solution which is fed to solvent extraction columns. Present processing methods remove the thin Zircaloy (trade name) coating which encases each rod by dissolution in an ammonium fluoride ammonium nitrate solution. The exposed uranium metal and fission products are then dissolved in a mixture of boiling nitric acid and aluminum nitrate. The metal dissolution reaction evolves NO_x such that the NO_x evolution rate is directly related to the metal dissolution rate. The reaction rate increases with:

- increasing acid, nitrate and dissolved uranium concentration
- increasing temperature
- increasing uranium metal surface area

As the metal rods dissolve, the solution ion concentrations, temperature and metal surface area change, causing the NO_x evolution rate to vary with time and creating peaks in the NO_x concentration of dissolver off-gases.

The fuel rod dissolution is carried out in three annular dissolvers which are maintained at a slight vacuum by individual off-gas systems. A schematic of a typical dissolver off-gas system is shown in Figure 3. Nitrogen oxides produced during dissolution are mixed with air before leaving the dissolver vessel via the downdraft condenser tower. The downdraft tower is a finned tube condenser which passes process vapors down

FIGURE 2
PLOT PLAN OF THE PUREX DISSOLVER AND MAIN BUILDING VENTILATION SYSTEMS

the shell side of the vessel and cooling water up through the tubes. Water and nitric acid vapors condense on the outside of the tubes, flow to the bottom of the condenser and back to the dissolver. In addition to vapor recovery, approximately 10 percent of the NO_x entering the tower is absorbed in the condensate and returned to the dissolver as nitric acid.

Dissolver off-gases leave the downdraft tower at approximately 30°C and pass through a tower packed with Pall rings. The packed tower functions as an ammonia scrubber during the coating removal process and is operated dry during metal dissolution. After leaving the ammonia scrubber, the dissolver off-gas is heated to approximately 200°C by a steam heater and electric heater in series. The off-gas then enters a second tower, or silver reactor, packed with Berl saddles. The tower packing is impregnated with silver nitrate which removes iodine from the process gases.

The dissolver off-gas is next filtered through two Fiberglass ^{Owens-} ~~Dow~~ Corning Trade name) filter bed units for removal of any remaining particulate materials. The primary filter, along with the dissolvers and connecting dissolver off-gas equipment, is located in a concrete cell within the main processing building. The back-up or secondary filter for each dissolver off-gas system is located in a vault, outside the main building (see Figure 2). Three parallel eight inch pipes, supported in a concrete encasement contain the off-gases as they flow from the main building to the back-up filter vault and on to the NO_x treatment facility.

Within the NO_x treatment facility, piping arrangements allow for the off-gas from one, two or all three dissolvers to be combined and fed to the treatment facility or by-pass the facility to be released via the stack. Off-gases from the dissolvers during the coating removal process are allowed to by-pass the facility since no NO_x is produced during this phase of operation. The NO_x from metal dissolution is routed through the two bubble-cap tray towers located within the treatment facility.

Off-gases pass through the two NO_x absorption columns at atmospheric pressure. Between 40 and 60 percent of the entering NO_x is absorbed in dilute nitric acid which is recycled from collection tanks located below the towers. Each tower contains eight trays with between 22 and 33 six-inch bubble caps per tray. Acid recycled to the first column (T-XA) is cooled by a heat exchanger. Additional liquid cooling is accomplished by routing inter-stage liquids from the top four plates to a second heat exchanger. Acid recycled to the second tower (T-XB) is cooled via a cooling coil installed within the acid collection tank. The acid recovered from the NO_x treatment facility is returned to the main building for concentration and reuse in the Purex process.

The off-gas is pulled from the dissolver, off-gas equipment and NO_x treatment facility by three large steam jets which may operate independently when the treatment facility is being bypassed, or in unison when pulling off-gases through the treatment facility. The off-gas then flows through three encased eight inch pipes to be discharged at the bottom of the 200 foot main ventilation stack.

FIGURE 3
DISSOLVER OFF-GAS SYSTEM

Treatment of Acidic Wastes

The Purex process produces an acidic waste stream which is processed to remove cesium and strontium, solidified and stored in tanks. The waste is accumulated in batches within the main processing building and treated in a processing step called sugar denitration. The sugar denitration process reduces the acid content of wastes before extracting cesium and strontium, resulting in reduced solid waste volumes to be stored in underground, double-shell tanks. The process requires the addition of sucrose to each batch of waste. The solution is heated to approximately 95°C and allowed to digest for 20 to 30 hours. Sucrose reacts with nitric acid in the waste to produce NO_x , carbon dioxide and water. The NO_x evolution rate is directly related to the rate of reaction which is dependent on:

- The sucrose concentration
- The acid concentration
- The metallic salt concentration
- The solution temperature

Each rate parameter listed above varies as the waste digests and the reaction proceeds, therefore, the NO_x concentration of the off-gas changes with time, producing peaks similar to peaks evolved from the uranium metal process.

A schematic drawing of the sugar denitration off-gas routing is shown in Figure 4. Sucrose is added to concentrated waste solutions in the first digestion tank (F15) and heated to approximately 95°C. Process off-gases are drawn through the tank vent to the waste concentrator where the gases combine with nitric acid vapors driven off during the concentration of dilute acid wastes. The NO_x and acid vapors flow through a demister to a bubble cap tray column where most of the acid vapor, but very little NO_x , is recovered. The off-gases are then routed through a condenser to a header system where they mix with gases vented from other process vessels.

After digesting in tank F15, the waste solution is transferred to a second digestion tank (F16). The denitration reaction is usually allowed to proceed to completion before transferring waste to tank F16. Premature transfers are sometimes required due to process upsets within the plant. Tank F16 is vented directly to the vessel vent system and no provisions are made to recover NO_x from the off-gas. Treated waste liquors are routed from tank F16 out of the main building to be collected in the AR vault.

Process and vessel vent off-gases are combined, routed through iodine and particulate control equipment to be mixed with air leaving the main building ventilation system. The combined gases are drawn through filters by three parallel fans, discharging to the main ventilation stack.

Minor Process Sources

The dissolution of Zircaloy coatings also removes small amounts of uranium

FIGURE 4
SUGAR DENITRATION OFF-GAS SYSTEM

from the fuel rods. To minimize uranium losses, coating wastes are processed, recovering a majority of the dissolved uranium. The recovery process converts uranium dissolved in the coating waste to uranium dioxide (UO_2) which is centrifuged and dissolved in nitric acid. The dissolution reaction produces NO_x at a rate dependent on the solution temperature and acid concentration; therefore, the NO_x evolution rate can be controlled by limiting the acid addition rate. Off-gases produced during the dissolution of recovered UO_2 are vented directly to the vessel vent system to be released via the main stack.

The Purex Plant will be provided with two processes for converting plutonium nitrate solutions to plutonium oxide powder. One of the processing methods, direct plutonium denitration, evolves small quantities of NO_x . Product solutions of plutonium nitrate are fed to a screw calciner where the solutions are heated, boiling off the acid and converting the nitrate to plutonium oxide. The conversion reaction produces NO_x . Calciner off-gases are scrubbed in a spray tower recovering nitric acid vapors while the remaining off-gas is released via the process vent system and main stack. No NO_x is recovered from the UO_2 dissolution or plutonium denitration off-gases.

Trace Sources

Trace emissions of NO_x result from:

- Nitric acid and uranium nitrate solution storage tank vents.
- Transfers of nitric acid from rail cars to storage tanks.
- Transfer of uranium nitrate solutions from storage tanks to tank trailers.
- Decontamination operations.

The listed sources are negligible when compared to process emissions and of intermittent duration, but, will contribute to the NO_x concentration of ambient air surrounding the Purex facility.

Quantification of Emissions

The NO_x composition of off-gas from the uranium metal dissolution and sugar denitration processes vary continuously (see previous sections). Therefore, instantaneous Purex Plant NO_x emissions will fluctuate around a daily average rate. The average NO_x emission rate is dependent upon the overall plant processing rate and whether the uranium metal and UO_2 dissolution processes are operating, while the magnitude of instantaneous NO_x emissions vary with:

- The number of batches of uranium metal dissolving simultaneously and the NO_x evolution rate from each dissolver.
- The number of batches of acidic waste denitrating simultaneously and the NO_x evolution rate from each tank.

- The NO_x evolution rate from the dissolution of UO₂.
- The NO_x evolution rate from the direct denitration of plutonium.

The dissolver off-gas system contains the only effective NO_x abatement equipment in the Purex Plant. This equipment is assumed to recover approximately 46 percent of the NO_x generated from the dissolvers under controlled conditions. Uncontrolled dissolver emissions were estimated assuming no recovery in the NO_x treatment facility. Since the off-gas systems of the sugar denitration, UO₂ dissolution and plutonium denitration processes contain no effective NO_x recovery equipment, the contribution of these processes to total Purex Plant NO_x emissions will not vary during controlled and uncontrolled operating conditions.

The four NO_x generating processes in the Purex Plant independently evolve NO_x at varying rates during different time periods each day. Table II lists estimates of the average daily emission durations and percent of total Purex Plant NO_x emissions evolved from each process under controlled and uncontrolled operating conditions.

TABLE II

Estimated Daily Duration and Percent of
Total Plant Emissions For Purex Processes Evolving NO_x

Process	Average Daily Duration of Process NO _x Emission, Hours	Percent of Total ⁽¹⁾ Daily Plant Emissions Evolved	
		Controlled Conditions	Uncontrolled Conditions
Uranium Dissolution	8	73	83
Waste Denitration	24	25	15
UO ₂ Dissolution	4	1	1
Plutonium Denitration	24	1	1

(1) Controlled conditions assume 46 percent of the NO_x entering the dissolver off-gas equipment is recovered which is equivalent to assuming normal operation of all Purex NO_x abatement equipment (downdraft tower and gas absorbers). Uncontrolled conditions assume no recovery of NO_x from dissolver off-gases. Under controlled operating conditions, the dissolution of uranium evolves 73 percent of the total daily plant NO_x emissions in an eight-hour period while a majority of the remaining daily emissions are spread out over a 24 hour period.

Plant operations allow a typical day to be divided into three time periods, during which emissions reflect different combinations of the four processes evolving NO_x. Instantaneous plant emissions will vary about a different

average value throughout each time period. Estimates of the duration and average NO_x concentration of stack effluents, assuming sequential operation of the uranium and UO_2 dissolution processes, are listed in Table III for controlled and uncontrolled conditions.

TABLE III

Estimated Fluctuation of Average Purex Plant NO_x Emissions

Time Period	Processes Assumed To Generate NO_x During Time Period ⁽¹⁾	Average Duration of Time Period, Hours	Average NO_x Concentration of Purex Plant Effluents ⁽²⁾	
			Controlled Conditions	Uncontrolled Conditions
A	UD + WD + PuD	8	800 ppm	1400 ppm
B	WD + UO_2D + PuD	4	110 ppm	110 ppm
C	WD + PuD	12	85 ppm	85 ppm
Time Weighted Daily Average	-	-	325 ppm	530 ppm

(1) UD = Uranium Dissolution, WD = Waste Denitration, UO_2D = UO_2 dissolution, PuD = Plutonium Denitration.

(2) Off-gas compositions were estimated assuming a total gas flowrate of 57,000 liters per second (121,000 cfm) at 40°C and 1 atmosphere. Controlled conditions assume that the dissolver off-gas abatement equipment recovers 46 percent of the entering NO_x while uncontrolled conditions assume no NO_x recovery.

Figure 5 shows the estimated variation of the average flowrate and NO_x concentration of off-gases entering the main Purex stack under controlled and uncontrolled conditions.

Peak NO_x evolution rates from the uranium dissolution and sugar denitration processes were estimated from past operating and laboratory data, modified to reflect flowsheet changes. Since the dissolution and denitration processes are operated independently, off-gases from different processes may peak simultaneously. A basis for worst case operating conditions was estimated to be represented by the simultaneous operation of:

- One dissolver at the average dissolution rate
- One dissolver at the peak dissolution rate
- One waste tank at the average digestion rate
- One waste tank at the peak digestion rate
- UO_2 dissolution and plutonium denitration at average rates

Time ⁽¹⁾ Period	Reference Point	Off-Gas Properties at Reference Point			
		Controlled Conditions		Uncontrolled Conditions	
		Flowrate, (2) Liters/Sec.	NO_x Concentration, PPM	Flowrate (2) Liters/Sec.	NO_x Concentration, PPM
A	1	330	121,500	380	197,500
	2	56,600	85	56,600	85
	3	57,000	800	57,000	1,400
B	1	-	0	-	0
	2	56,600	110	56,600	110
	3	56,600	110	56,600	110
C	1	-	0	-	0
	2	56,600	85	56,600	85
	3	56,600	85	56,600	85

(1) Time period durations and operating processes defined in Table III.

(2) Assumed off-gas conditions: 40°C and 1 atmosphere. Controlled conditions assume that dissolver off-gas abatement equipment recover 46% of the entering NO_x while uncontrolled conditions assume no NO_x recovery.

FIGURE 5

ESTIMATED VARIATION OF AVERAGE NO_x CONCENTRATION AND FLOWRATES FOR OFF-GASES ENTERING THE MAIN PUREX STACK

Assuming the above basis, a total off-gas flowrate of 56,600 liters per second at 1 atmosphere and 40°C and normal operation of the Purex NO_x abatement equipment, the Purex main stack effluents are estimated to reach an instantaneous peak NO_x concentration of 4700 ppm. The peak conditions represent a theoretical maximum NO_x emission rate and will not generally occur unless a number of operating problems are encountered simultaneously.

Annual NO_x emissions from the Purex Plant are directly related to the amount of fuel processed each year. Estimates of controlled and uncontrolled emissions for the present projected production schedule are listed by year in Table IV.

TABLE IV
Annual Purex Plant NO_x Emissions

Calendar Year	Total Uranium Processed, Metric Tons	Annual NO _x Emissions, ⁽¹⁾ in Metric Tons of NO ₂	
		Controlled Conditions	Uncontrolled Conditions
1981	517	145	238
1982	856	240	394
1983	1026	288	472
1984	688	193	316

(1) Controlled conditions assume 46 percent of the NO_x entering the dissolver off-gas equipment is recovered. Uncontrolled conditions assume no recovery of NO_x from dissolver off-gases.

Operating periods for the Purex Plant will average approximately six months per year, including time for start-up, mechanical failures and operating difficulties. The plant will emit NO_x only during the time periods when fuel is being processed. On-line processing periods are estimated to last a maximum of 112 days per year. The impact of Purex Plant NO_x emissions on the ambient air quality will be predicted assuming a daily time averaged effluent of 57,000 liters per second containing 325 ppm NO_x and lasting 112 days per year. The opacity of the off-gas plume is expected to exceed emission standards due to the presence of NO₂.

AR VAULT

Acidic wastes from the Purex process are accumulated in two tanks within AR Vault. The accumulated batches of waste are then used as feed solutions for the waste fractionization facility where cesium and strontium are recovered. The stored waste contains dilute nitric acid, therefore, the tank ventilation air will contain small quantities of NO_x.

The ventilation system for acid waste storage tanks in the AR Vault is shown in Figure 6. Vapors from the storage tanks are collected in a vent header. The off-gas is then heated, filtered through a three stage filter and prefilter, reheated and passed through a second three stage filter. The off-gas system is maintained at a negative gauge pressure by a fan or emergency air jet which exhaust into a stack, 150 feet high and 8 inches in diameter. A worst case estimate of AR vault NO_x emissions is listed in Table V.

TABLE V

Estimated AR Vault NO_x Emissions

Total Off-Gas Flowrate ⁽¹⁾	310 liters/second
Maximum NO_x Concentration	18 ppm
Annual Total NO_x Emissions	0.36 Metric Tons NO_2

(1) Assumed Off-gas Conditions: 1 atmosphere and 30°C

The impact of AR Vault emissions on the ambient air quality is predicted by assuming a continuous stack off-gas of 310 liters per second containing 18 ppm NO_x for 365 days per year. The opacity of AR Vault emissions will comply with emission standards during normal operating conditions.

THE UO_3 PLANT

Uranium nitrate product solutions from the Purex process are transported by truck to the UO_3 Plant for conversion to uranium trioxide (UO_3) powder. The UO_3 is then packaged and shipped to off-site locations for enrichment and reuse as reactor fuel. Two UO_3 Plant processes evolve NO_x , concentration of the uranium nitrate feed solution and calcination of the concentrated feed to UO_3 powder.

Feed Concentration

The initial step in processing uranium nitrate solutions at the UO_3 Plant is feed concentration. Feed liquors from the Purex Plant are heated in one of three concentrators, driving off water, nitric acid vapor and small amounts of NO_x . A sketch of the UO_3 Plant off-gas system is shown in Figure 7. Off-gases leave each concentrator through a knock-out pot and are combined in a vent header. The off-gases then pass through a catch tank, condenser and steam jet before combining with the off-gases from the uranium calciners.

Uranium Calcination

Concentrated uranium solution is fed to one of six stirred bed calciners. The solution is heated to approximately 270°C to form UO_3 while liberating NO_x , water and oxygen. Off-gases from each calciner are passed through a

FIGURE 6
AR VAULT OFF-GAS SYSTEM

FIGURE 7
110. PLANT OFF-GAS SYSTEM

wet scrubber which removes entrained UO_3 powder, cools the off-gas and absorbs a small amount of NO_x . The vapors from each calciner are then combined in a vent header and air added before routing the off-gas to a gas cooler.

The gas cooler consists of a finned tube heat exchanger with process vapor on the shell side and cooling water on the tube side. Process vapors are cooled to 30°C , condensing out water which runs down the fins and outer tube walls, absorbing approximately 40 percent of the NO_x entering the cooler. After leaving the gas cooler, the off-gases enter an acid absorption tower. The tower contains 20 bubble cap trays with cooling coils on the bottom 11 trays and operates at atmospheric pressure. NO_x is absorbed by dilute nitric acid to form a product which is approximately 50 weight percent acid. A steam jet pulls the gases through the calciner off-gas system and combines the gases with vapors from the feed concentrators. The combined vapors are passed through a condenser and mixed with air before exhausting to a stack, 80 feet high and 10 inches in diameter.

Trace Sources

Trace emissions of NO_x will be evolved from sources identical to the trace sources identified for the Purex Plant. The NO_x emitted from these sources will contribute to the NO_x concentration of ambient air surrounding the UO_3 Plant, but, will be negligible in comparison to process emissions.

Quantification of Emissions

The UO_3 Plant will produce an off-gas stream of constant composition for approximately 30 days per year. Controlled NO_x emissions from the UO_3 Plant are estimated from overall material balances and past operating data, indicating an overall UO_3 Plant NO_x recovery efficiency of 93 to 97 percent. Uncontrolled emissions are estimated assuming no NO_x is recovered from the plant off-gases, representing the failure of the gas cooler - absorption tower system. Estimates of controlled and uncontrolled UO_3 Plant stack emissions are summarized in Table VI.

TABLE VI

UO_3 Plant NO_x Emissions

	<u>Operating Conditions</u> ⁽¹⁾	
	<u>Controlled</u>	<u>Uncontrolled</u>
Flowrate, Liters/second ⁽²⁾	1900	1,900
NO_x Concentration, ppm	3000	42,000

⁽¹⁾ Controlled emissions assume an overall plant NO_x recovery efficiency of 93 percent.

⁽²⁾ Assumed off-gas conditions: 1 atmosphere and 30°C .

Total annual UO_3 Plant NO_x emissions will depend upon the Purex Plant production schedule. Table VII lists controlled and uncontrolled annual emissions from the UO_3 Plant assuming the present projected processing schedule.

TABLE VII

Annual UO_3 Plant NO_x Emissions

Calendar Year	Uranium Processed, Metric Tons	Annual NO_x Emissions, ⁽¹⁾ in Metric Tons of NO_2	
		Controlled Conditions	Uncontrolled Conditions
1982	686	19	266
1983	1,372	38	532
1984	680	19	266

(1) Controlled emissions assume an overall plant NO_x recovery efficiency of 93 percent.

The impact of UO_3 Plant NO_x emissions will be estimated assuming a stack effluent of 1,900 liters per second containing 3,000 ppm NO_x for a duration of 30 days per year. Observations by operating personnel indicate that the resultant plume opacity will exceed the State and local opacity standard.

Power Plants

Two coal burning power plants, one located in each of the 200 areas, produce process steam for the Purex and UO_3 Plants. The east area powerhouse provides steam to the Purex Plant while the west area powerhouse supplies the UO_3 Plant. Annual and instantaneous emissions from the power plants, with and without the reprocessing facilities operating, are listed in Table VIII. By fiscal year 1981, pollution abatement scrubbers are proposed to be in operation, reducing particulate and sulfur oxide emissions from the power plant stacks. No significant changes in the rate of powerhouse NO_x emissions are expected after installation of the new equipment. The impact of power plant NO_x emissions on the ambient air quality is predicted by assuming an incremental increase in the powerhouse NO_x emission rate of 35 ppm NO_x at a flowrate of 69,800 liters per second for 365 days per year. Since the major component of powerhouse NO_x emissions is in the form of colorless nitric oxide, changes in the powerhouse NO_x emission rates will not affect the opacity of powerhouse plumes.

TABLE VIII

Power Plant NO_x Emissions

	Annual Coal Consumption, Metric Tons	Off-Gas ⁽¹⁾ Flowrate, Liters/Second	NO _x Concentration, PPM	Total Annual NO _x Emissions, Metric Tons
Reprocessing Facilities not Operating ⁽²⁾	93,000	69,800	375	700
Reprocessing Facilities Operating	100,000	69,800	410	760

(1) Combines off-gases from both east and west powerhouses.
Assumed off-gas conditions: 1 atmosphere and 200°C.

(2) Annual average of 1977 and 1978.

IMPACT OF 200 AREA NO_x EMISSIONS ON THE AMBIENT AIR QUALITYOCCUPATIONAL AREAS

Nitrogen dioxide (NO₂) concentrations in the air near the Purex Plant were measured during February, 1968 through June, 1968 by Battelle Pacific Northwest Laboratories⁽³⁾ while the plant was operating. NO_x in ambient air is measured as NO₂ since this is the predominant form found under ambient conditions ($2\text{ NO} + \text{O}_2 \rightarrow 2\text{ NO}_2$, at ambient temperatures). Measurements were obtained from three separate locations: 1) A site approximately one mile southeast of the Purex Plant, 2) a site at the southeast corner of the 200 East Area, and 3) a site directly east of Purex. The sample locations are illustrated in Figure 8. A wind rose, typical of the Hanford Reservation, is included to demonstrate the sample locations with respect to the predominant winds. Sources, besides Purex, which influence NO_x concentrations at these locations are the AR Vault and the East Area powerhouse.

During the four and one-half month study, background levels of 0.012 part per million (ppm) NO₂ were observed 90 percent of the time. Levels above background never exceeded 0.27 ppm. These concentrations fall well below the occupational Threshold Limit Value (TLV) for an eight-hour day of 5.0 ppm. NO_x emissions, following the resumption of fuel reprocessing in the 200 Areas, are expected to remain at those levels experienced during previous operating periods. Therefore, NO_x concentrations in the vicinity of processing facilities are expected to be similar to those previously reported near Purex.

Typical Hanford Wind Rose

Figure 3

Ambient Air Monitoring Site Locations Near The Purex Plant, February to June, 1968.

Studies on the effect of low concentrations of NO_x on human health have not agreed upon a specific concentration at which adverse effects occur. Findings of the World Health Organization (WHO) indicate that increases in respiratory illness result from intermittent exposures of 0.47 ppm NO_2 . This concentration was selected by WHO as the lowest NO_2 concentration at which adverse health effects might be expected (4). The American Lung Association (ALA) report change in ventilatory function and increase in respiratory illness occur at concentrations as low as 0.15 ppm NO_2 (5). Further studies indicate that no association exists between continual exposure to ambient NO_2 concentrations, ranging from 0.053 ppm to 0.30 ppm, and increases in respiratory diseases (6).

NO_x concentrations in the vicinity of the Purex Plant during past operations did not result in any noticeable adverse health effects on workers. Similarly, future NO_x concentrations near Purex due to operations are not expected to pose any significant health hazards.

NONOCCUPATIONAL AREAS

Past Measurements

Offsite ambient air NO_x concentrations (measured as NO_2) have been determined and reported quarterly by the Hanford Environmental Health Foundation (HEHF). Ambient air measurements were made both while Purex was operating (1969, 70, 71, and 72) and during the years following its shutdown (1973, 74, and 75). Several sampling locations were located opposite the project boundary in the predominantly downwind direction from Purex (see Figure 9). A wind rose demonstrates the relationship between the sampling locations and the predominant wind conditions.

The quarterly average concentrations reported by HEHF are illustrated in Table IX (7). Only the Richland, North Richland, and 300 Area locations were sampled throughout the period of interest.

The annual average concentrations were calculated from the quarterly averages and are presented in Table X. Annual averages ranged from 0.0076 ppm to 0.0016 ppm, all well below the national ambient air standard for NO_2 of 0.05 ppm, annual arithmetic mean. A comparison of annual averages for the individual sites during and following Purex operations yields no trends or significant differences, demonstrating that Purex operations had no measurable influence on offsite concentrations.

Figure 9.

Hanford Area Ambient Air Monitoring Sites, 1969 to 1975.

TABLE IX

Quarterly Average NO₂ Concentrations, ppm⁽¹⁾

Date	Opposite ⁽²⁾ Richland	Opposite ⁽²⁾ N. Richland	Opposite 300 Area	Ringold ⁽³⁾	White ⁽³⁾ Bluffs
During Operations					
1969 1st Qtr.	-	-	0.0020	0.0017	0.0017
2nd Qtr.	-	-	0.0013	0.0018	0.0016
3rd Qtr.	-	-	-	-	-
4th Qtr.	-	-	< 0.0023	< 0.0024	0.0015
1970 1st Qtr.	0.0036	0.0052	0.0020	< 0.0020	0.0014
2nd Qtr.	0.0026	0.0028	0.0020	0.0020	0.0027
3rd Qtr.	0.0130	0.0100	0.0070	0.0090	0.0100
4th Qtr.	0.0090	0.0080	0.0080	0.0080	0.0070
1971 1st Qtr.	0.0070	0.0050	0.0060	0.0060	0.0060
2nd Qtr.	0.0030	0.0030	0.0040	0.0050	0.0050
3rd Qtr.	0.0070	0.0080	0.0090	0.0100	0.0090
4th Qtr.	0.0040	0.0030	0.0040	0.0040	0.0040
1972 1st Qtr.	< 0.0050	< 0.0020	< 0.0010	< 0.0030	< 0.0010
2nd Qtr.	0.0050	0.0020	0.0030	0.0020	0.0040
3rd Qtr.	0.0060	0.0060	0.0050	0.0050	-
4th Qtr.	0.0090	0.0060	0.0060	0.0070	-
Following Operations					
1973 1st Qtr.	0.0037	0.0055	0.0050	0.0100	-
2nd Qtr.	0.0070	0.0060	0.0050	0.0060	-
3rd Qtr.	0.0090	0.0110	0.0120	0.0050	-
4th Qtr.	-	0.0080	0.0060	-	-
1974 1st Qtr.	0.0040	0.0040	0.0030	-	-
2nd Qtr.	-	0.0050	0.0060	-	-
3rd Qtr.	0.0070	0.0070	0.0050	-	-
4th Qtr.	0.0050	0.0060	0.0050	-	-
1975 1st Qtr.	0.0030	0.0040	0.0030	-	-
2nd Qtr.	0.0030	0.0030	0.0030	-	-
3rd Qtr.	0.0050	0.0050	0.0040	-	-
4th Qtr.	0.0050	0.0050	0.0040	-	-

(1) Detection level of 0.001 ppm

(2) Sampling initiated 1970

(3) Sampling discontinued 1973

- Indicates no sample taken

TABLE X

Annual Average NO₂ Concentrations, ppm

Year	Opposite Richland	Opposite N. Richland	Opposite 300 Area	Ringold	White Bluffs
1969	-	-	0.0019	0.0020	0.0016
1970	0.0070	0.0065	0.0048	0.0053	0.0053
1971	0.0053	0.0048	0.0058	0.0063	0.0060
1972	0.0063	0.0040	0.0038	0.0043	0.0025
1973	0.0066	0.0076	0.0070	0.0070	-
1974	0.0053	0.0055	0.0048	-	-
1975	0.0040	0.0043	0.0035	-	-

Recent Measurements

The collection and analysis of ambient air samples for NO_x concentrations was discontinued in 1975 since it appeared that no measureable changes were occurring over the years. During July 1978, in conjunction with the Purex preoperational surveillance program, the measurement of ambient NO_x concentrations was reinitiated. Figure 10 shows the sampling locations chosen to best evaluate the environment around Purex prior to and following its start-up. Average NO₂ concentrations (ppm) observed at these locations during July 1978 through August 1978 are given in Table XI below:

TABLE XI

Average 200 Area Ambient Air NO_x Concentrations,
July and August, 1978 (1)

<u>Monitoring Site Location</u>	<u>Average Ambient NO_x Concentration, ppm</u>
Sullivan Barn	<0.005
ALE Laboratory	<0.004
Rattlesnake Springs	<0.004
100-B Area	<0.004
100-D Area	<0.003
615 Building	<0.004
Army Barracks	<0.004
FFTF	<0.004

(1) Detection level = 0.003 ppm

Average concentrations are reported as "less than" values since some of the individual samples taken during this time period were below the instrument detection level and were reported as "less than" numbers. All the recently determined average concentrations range between <0.003 ppm and 0.005 ppm. These concentrations fall within the range of concentrations reported during 1969 through 1975.

Predicted Incremental Impact of Reprocessing Operations

The atmospheric dispersion of pollutants is commonly described mathematically by a normal distribution model. Dispersion coefficients explaining the lateral and vertical diffusion of the plume are estimated using one of several methods. Using the following model, ground level concentrations for various locations can be predicted.

$$x = \frac{Q}{\pi \sigma_y \sigma_z u} \exp \left[-\frac{y^2}{2\sigma_y^2} - \frac{h^2}{2\sigma_z^2} \right]$$

where,

x = ground level concentration, ppm

Q = source strength, ppm (m^3/sec)

π = pi, 3.1416

y = crosswind distance measured horizontally from centerline of cloud, m

σ_y = crosswind lateral standard deviation of cloud concentration, m

σ_z = crosswind vertical standard deviation of cloud concentration, m

u = wind speed in direction of travel, m/sec

h = height of release, m

from: Air Pollution Control, Environmental Science and Technology, Wiley-Interscience, 1971. (8)

Assuming that we are interested only in centerline concentrations and that the stack height is the effective plume height, both worst case assumptions, the dispersion model equation is further simplified to:

$$x = \frac{Q}{\pi \sigma_y \sigma_z u} \exp \left[-\frac{h^2}{2\sigma_z^2} \right]$$

Using the above model with dispersion coefficients derived from Pasquill's curves (8), ground level concentrations were predicted for six locations around the Hanford Reservation. These locations are: (see Figure 10 for map)

- 1) Opposite 300 Area
- 2) Opposite Richland
- 3) ALE - Rattlesnake Mountain
- 4) Yakima Barricade
- 5) White Bluffs
- 6) Ringold

In determining the theoretical NO_x concentrations due to Purex operations at these locations, several assumptions were made. Where practical, worst case conditions were assumed in order to ensure that conservative values were predicted. That is, predicted values should be higher than actually observed in the field. These assumptions are listed below:

- effective plume height equals stack height
- interested only in centerline concentrations
- constant emission rates from stack

Once concentrations were predicted for a source over all the atmospheric stability categories, an average concentration was calculated using the percentage of time each stability generally exists per year over the Hanford Reservation. Next, the annual contribution to a certain location was calculated based on what percentage of time the general wind direction is towards the site of concern. Finally, the annual contribution of NO_x to the annual average ambient concentration at each location was further reduced based on the amount of time the various plants were actually operating during the year.

Results of the prescribed calculations are reported in Table XII for those facilities with NO_x emissions which are attributable to the startup and operation of Purex.

TABLE XII

Predicted Incremental Contributions to Non-Occupational
Ambient Air NO_x Concentrations, ppm

	Purex	UO ₃ Plant	Powerhouse	AR Vault
Opposite 300 Area	0.0010	0.0001	0.0005	negligible
Opposite Richland	0.0010	0.0001	0.0005	"
ALE - Rattlesnake Mtn.	0.0010	0.0001	0.0004	"
Yakima Barricade	0.0008	0.0001	0.0004	"
White Bluffs	0.0008	0.0001	0.0003	"
Ringold	0.0010	0.0001	0.0005	"

FIGURE 10
Hanford Area Ambient Air Sampling Sites,
July and August 1978

NO_x emissions from the AR Vault are considered negligible since calculations showed concentrations at the site boundary of 1.0×10^{-6} ppm or less. The total effect of the 200 Area NO_x emissions on the offsite ambient air quality was calculated by combining all the sources for each given location. (see Table XIII).

TABLE XIII

Hanford Reprocessing Facilities Contribution to
Non-Occupational Ambient Air NO_x Concentrations

<u>Monitoring Site Location</u>	<u>Ambient NO_x Concentration, ppm</u>
Opposite 300 Area	0.002
Opposite Richland	0.002
ALE - Rattlesnake Mtn.	0.002
Yakima Barricade	0.001
White Bluffs	0.001
Ringold	0.002

Based on the theoretical calculations, the largest increase to the offsite ambient air NO_x concentration is 0.002 ppm on an annual average basis. Present ambient air NO_x concentration levels are less than 0.004 ppm. Therefore, the non-occupational ambient air NO_x concentration will not be increased to significant levels by reprocessing operations in the Hanford 200 Areas.

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