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Project Summary

A Model for Evaluation of Refinery and Synfuels VOC Emission Data

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Estimates of the emissions of volatile organic compounds (VOCs) from petroleum refineries and synfuel plants are of considerable interest to EPA, industry, and the public. Such estimates are needed in the preparation and review of Environmental Impact Statements (EIS) and permits required by the Clean Air Act. In response to this need, several studies have been made of VOC emissions, particularly from refineries. Methods for estimating VOC emissions and the results of VOC emissions tests have been published in various journals and at numerous forums. A need has developed to define a consistent and comprehensive approach for estimating VOC emissions from refineries and synfuel plants.

This study has resulted in the development of a model for performing such estimates. A modular technique was developed in which the entire spectrum of potential VOC emissions sources was defined in a number of process and utility modules. Each module represents a process or auxiliary unit. The user of the model provides emission source counts and other process information, or uses default values provided. Emissions are calculated, using emission factors for each source type. Detailed examples of the application of the model to both refineries and synfuels plants are presented.

This Project Summary was developed by EPA's Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in two separate volumes (see Project Report ordering information at back).

Introduction

Over the past several years, volatile organic compound (VOC) emissions from petroleum refineries and synfuel plants have been of considerable interest to the EPA, industry, and the general public. The preparation and review of Environmental Impact Statements (EIS) and permitting requirements of the Clean Air Act depend on emission estimates. In response to this need, several studies have been made of VOC emissions, particularly from refineries. Methods for estimating VOC emissions and the results of VOC emissions tests have been published in various journals and at numerous forums. A need has developed to define a consistent and comprehensive approach for estimating VOC emissions from refineries and synfuel plants. This study was performed to fulfill this objective.

A literature search was conducted to obtain all available information on VOC emissions from petroleum refineries and synfuel plants. The types of synfuel plants included in the search were coal gasification (excluding in-situ gasification), coal liquefaction (direct and indirect), and oil shale processing.

Four major sources of emissions were included in the search: process emissions, product storage, baggable fugitive emissions, and nonbaggable fugitive emissions. Both controlled and uncontrolled sources were considered; if the source was controlled, any available information on the degree and type of control and the rationale for control application was included.

Process operating parameters and physical data were included if they pertained to a process stream for which emission data were expected to be available or if they pertained to any existing emission model. Emission data could include measurements of emission rates, measurements of parameters that could correlate with or predict emission rates, or composition data.

Because of Radian's involvement with EPA in VOC emissions activities over the past 7 years, it was expected that very little information of significance would be found of which EPA and Radian were not already aware. A search of the DOE ENERGY data base using the DIALOG Information Retrieval Service bore this out. The search included the last 5 years. Therefore, the bulk of the information was gathered through the Radian library. Particularly in the refinery area, a great deal of the available information on emissions is the result of EPA/Radian testing efforts.

Refinery emission data were obtained from a few major sources which had been identified from past studies. These sources are tabulated in the full report. Some of these references also provided additional data (e.g., emission source distributions and process and operating parameters) needed to develop a model for estimating refinery VOC emissions.

Much less information on VOC emissions from synfuels plants is available than for refineries. The full report summarizes the literature surveyed. Source types and frequencies for a number of synfuel processes, together with a limited amount of emission factor data (primarily for Lurgi gasification plants), were located.

There are thousands of potential VOC emission sources in a refinery or synfuel plant, but this variety of sources falls into one of the following general categories:

- *Process fugitive emissions*. These are the result of leakage of VOC from the piping and fittings with which a process unit is constructed. Sources of process fugitive emissions and their uncontrolled emission factors are given in Tables 1 and 2. Note that the emission factors are presented by industry, and that there are significant differences between industries. The full report describes how VOCs may be emitted from each source type, how such emissions may be controlled, and the effectiveness of these control measures.

Table 1. *Process Fugitive Emission Factors*

Source Type	*Service Category*	*Emission Factors, lb/day/source*	
		Refineries	*SOCMI*[a]
Pump Seals	*Light Liquid*[b]	*6.0*	*2.6*
Pump Seals	*Heavy Liquid*[c]	*1.1*	*1.1*
Compressor Seals	*Hydrocarbon Gas*	*34.0*	*12.0*
Compressor Seals	*Hydrogen*[d]	*2.6*	*–*
Valves	*Hydrocarbon Gas*	*1.4*	*0.30*
Valves	*Hydrogen*	*0.43*	*–*
Valves	*Light Liquid*	*0.58*	*0.38*
Valves	*Heavy Liquid*	*0.012*	*0.012*[e]
Connections	*All*	*0.013*	*0.044*
Relief Valves	*Gas*	*8.6*	*5.5*
Relief Valves	*Liquid*	*0.37*	*0.37*[e]
Open End Lines	*All*	*0.12*	*0.09*
Process Drains	*All*	*1.7*	*–*

[a]The Synthetic Organic Chemical Manufacturing Industry. These emission factors may be more appropria for petrochemical units associated with refineries or synfuel plants.
[b]Any organic material more volatile than kerosene.
[c]Any organic material with a volatility equal to or less than kerosene.
[d]A stream with greater than 50 percent (by volume) of hydrogen.
[e]From refinery data since there were not enough heavy liquid sources found in the SOCMI testing to warra the development of separate emission factors.

- *Process combustion emissions*. Many refinery or synfuel processes require a great deal of heat input, which may be provided directly by a fixed process heater, or indirectly by steam, generated in a boiler. Incomplete fuel combustion and/or reactions between the products of combustion may result in VOC emissions. Emission factors from combustion sources are given in Table 3.
- *Process point source emissions*. Point sources of VOC emissions are present in some process units, and emissions must of necessity be estimated for each individual process unit. Data obtained in this study were used to identify the point sources occurring in various process units and to develop emission factors for each.
- *Blowdown and flare system emissions*. Flares are used to handle large emergency releases from refinery and synfuel plant process units and for combusting continuous, low flows of VOC that are trans ported in closed vent systems. Flar destruction efficiencies may rang from 91 to 100 percent; a mean eff ciency of 98 percent is normally as sumed.
- *Wastewater treatment system emis sions*. Primary sources of VO emissions from wastewater treat ment systems are evaporative emis sions from oil/water separators an dissolved air flotation units. Con trolled and uncontrolled emissio factors are given in Table 4.
- *Sludge/solid waste treating emis sions*. Atmospheric VOC emission can result from the land disposal o refinery and synfuel plant oil wastes. No well-established emis sion factors exist for any of the im portant disposal methods (land farming, landfilling, and surfac impoundment), but the full repor presents several predictive emis sion models which have been pro posed in the literature.

Table 2. *Process Fugitive Emission Factors Used in the Gasification, Acid Gas Removal, and Wastewater Extraction Modules*

Source Type	*VOC Emission Factor, lb/day/source*[a]
Pump Seals - Aqueous	*0.0026*
Pump Seals - Hydrocarbon Liquid	*0.011*
Compressor Seals - Hydrocarbon Gas	*34.0*
Compressor Seals - Hydrogen Gas	*2.6*
Valves - Hydrocarbon Gas	*0.0042*
Valves - Hydrogen Gas	*0.43*
Valves - Hydrocarbon Liquids	*0.0057*
Valves - Aqueous	*0.0026*
Connections - Hydrocarbon Gas	*0.0005*
Connections - Hydrocarbon Liquid	*0.0011*
Connections - Aqueous	*<0.00007*
Relief Valves - Gas	*0.34*
Relief Valves - Liquid	*0.0037*
Open End Lines	*0.12*
Process Drains	*1.7*
Sample System Purging	*0.79*

[a]*There is some concern over the accuracy of these numbers, since they represent only the gaseous portion of the leak (i.e., they do not include the potential contribution of liquid leaks). A number of liquid leaks were noted, although most were in aqueous stream service. These factors were included because they are the only source of gasification specific data, but the use of refinery factors may be more accurate if liquid leaks are suspected to be significant.*

- *Emissions from storage tanks.* Emission models have been developed for the most commonly used types of tanks used to store crude oil and liquid products or byproducts. These models are quite complex; details are given in the full report.
- *Emissions for cooling towers.* VOC emissions from cooling towers typically occur as a result of leaks in shell-and-tube heat exchangers through which cooling water circulates. An emission factor of 6 lb VOC/10^6 gal. of cooling water circulated is used.
- *Emissions from product loading operations.* VOC emissions result from evaporation of products during loading operations. Emission factors for several different stocks, means of transport, and style of loading are given in Table 5. For other products, emissions may be estimated by using the factors for the product listed whose volatility is closest to the product of interest.

Table 3. *Emission Factors for Heaters and Boilers*

	Emission Factors, lb/10^6 Btu	
Fuel Type	*Industrial Heaters and Boilers ($<100 \times 10^6$ Btu/hr)*	*Utility Boilers ($\geq 100 \times 10^6$ Btu/hr)*
Natural Gas	*0.0029*	*0.001*
Fuel Oil	*0.0667*	*0.0667*
Coal (Bituminous or Lignite)	*1.0 lb/ton*	*0.3 lb/ton*
Coal (Anthracite)	*negligible*	*negligible*

Refineries and synfuel plants may be thought of as consisting of a number of process units and auxiliary operations. To provide a VOC emission model, a number of such process and auxiliary units were selected. Process and auxiliary modules were developed to represent the process units and auxiliary operations in their generic form. Modules were assigned to those processes which may potentially make a significant contribution to VOC emissions. The refinery and synfuel modules considered in the VOC emission model developed as a result of this study are listed in Tables 6 and 7. Note that there is some overlap; a number of the refinery modules will be found in most synfuel plants. The full report describes each module so that the user may select those which are applicable to his refinery or synfuel plant.

Information on the numbers and types of VOC emission sources occurring in each module was used to develop various levels of default values. These defaults provide useful information to users of the model who may have different amounts of detailed data regarding a specific refinery or synfuels plant for which an estimate of VOC emissions is desired.

Results

The VOC emission model is presented in a workbook format in appendices to the full report. The model consists of calculation sheets and module default sheets. The basic emission calculations for all emission sources are done on the calculation sheets. If the person using the model has complete descriptive information about the plant in question, then the calculation sheets will provide everything else necessary to estimate the VOC emissions. In most cases, however, the calculation sheets will require some input data that the user does not have, and the default sheets were designed to provide reasonable estimates for such missing data.

The logic flow of the emission model is illustrated in Figure 1. The user first characterizes the plant to be modeled by selecting appropriate process and auxiliary modules. Process modules are the model's representation of process units (such as a Fluid Catalytic Cracker, a Naphtha Hydrotreater, or a Lurgi Gasifier). Auxiliary modules are the representation of non-process operations (such as wastewater treating, cooling towers, and product storage). If the user does not know which modules should be included, several typical refineries and synfuel plants are fully defined. These "generic plants" may be used as is or simply as a guide in selecting the modules for a particular plant.

The emissions are calculated on a module-by-module basis, using emission calculation sheets and default sheets (as necessary). When all the process modules have been calculated, a similar procedure is followed for the auxiliary modules. The results may be displayed in at least two useful ways. First, the emission estimates on a module-by-module basis will show which modules are producing the most emissions; control efforts can be concentrated where they will accomplish the greatest emissions reductions. Second, adding together the emissions from like sources (e.g., light liquid pump seals) can facilitate comparisons of potential reductions which may be achieved by control programs aimed at all sources of a given type, such as leak detection and repair programs or improved equipment specifications.

Several examples of the use of the VOC emission model are detailed in the full report. One of the example plants was a small refinery. Table 8 lists the modules used to represent the small refinery, and Figure 2 is a block diagram. The results of the model VOC estimate are summarized in Table 9. As described previously, the VOC model has multiple levels of defaults to allow the user to take advantage of whatever data is available. Table 10 compares the model results, using three levels of defaults.

Conclusions and Recommendations

This report presents a mathematical model for estimating VOC emissions from refineries and several types of synfuel plants. All significant VOC emission sources have been included in the emissions model. A modular technique was developed in which the entire spectrum of potential VOC emissions sources was defined in a distinct number of process and utility modules. This model is convenient, flexible, and functional for developing VOC emissions estimates for very diverse petroleum refineries and synfuel plants.

The model developed in this study

Table 4. *Emission Factors for Wastewater Treating Blowdown Systems, Flares, and Cooling Towers*

Source Type	Emission Control	Emission Factor
Wastewater Treating		
Oil/Water Separator	Uncovered	1.88 lb/10^3 gal. WW
Oil/Water Separator	Covered	0.38 lb/10^3 gal. WW
Oil/Water Separator	Covered and vented to flare	0.06 lb/10^3 gal. WW
Dissolved Air Flotation	NA	0.09 lb/10^3 gal. WW
Blowdown and Flares	NA	0.8 lb/10^3 bbl crude
Cooling Towers	NA	6 lb/10^6 gal. CW

Table 5. *Emission Factors for Product Loading*

		Emission Factor, lb/10^3 gal.				
Vehicle	Loading Style	Gasoline	Jet Naphtha	Kerosene	No. 2 Fuel Oil	No. 6 Fuel Oil
Tank Trucks/ Tank Cars	Submerged-normal	5.0	1.5	0.02	0.01	0.0001
	Splash-normal	12.0	4.0	0.04	0.03	0.0003
	Submerged-balanced	8.0	2.5	NA	NA	NA
	Splash-balanced	8.0	2.5	NA	NA	NA
Barges	Clean-vapor free	1.2				
	Uncleaned-dedicated	4.0				
	Average condition	4.0	1.2	0.13	0.012	0.00009
Ocean Barges	Clean-vapor free	1.3				
	Uncleaned-dedicated	3.3				
	Ballasted	2.1				
Marine Tankers	Clean-Vapor free	1.0				
	Ballasted	1.6				
	Uncleaned-dedicated	2.4	0.5	0.005	0.005	0.00004
	Average condition	1.4				

has several unique and valuable features. The modules lend themselves readily to individual updating, improvement, and expansion, without disturbing the integrity of the remaining modules. The model is capable of developing emissions estimates from various levels of information. In the extreme, VOC emission estimates for refineries and synfuel plants can be developed when only the plant type and capacity are known. The results of these "maximum default" cases are presented as Table 10.

Several areas for further work could enhance the model developed in this study. The most obvious is computerization of the model. The modular form of this model is ideal for computerization. A computerized version of the model would allow rapid estimation of VOC emissions and optimization of processing and control techniques for minimizing VOC emissions. Different levels of control could be quickly evaluated under different scenarios. Summaries of emissions from particular sources across modules could be prepared with minimal effort.

VOC emissions from fugitive process sources (valves, pumps, flanges, etc.) represent a significant percentage of total VOC emissions. Emissions from these sources are best controlled by a leak detection and repair program. This VOC emissions model could ultimately incorporate EPA's leak detection (LDAR) model to allow additional evaluation and emission minimization studies to be performed rapidly. The LDAR model is currently in computer form.

The accuracy of the VOC emission estimate is not evaluated by the current model. An assessment of accuracy would require information on the accuracy of the emission source data as well as the equipment counts and loading levels. This information is available (in the form of confidence intervals, standard errors, and other types of error bounds) for some of the data used in developing the model. For other sources, new data are currently being developed which should include an accuracy assessment. The level of accuracy in using the model will also depend on the level of information that the user has available (e.g., equipment counts versus unit capacity levels). The current model could be updated to include levels of accuracy for all default values. These values could then be summarized by appropriate error propagation meth-

Table 6. *Refinery Modules*

Module Name	*Comments*
1. Atmospheric Crude Distillation	*Includes desalting, heat exchange network, atmospheric column, and side stream strippers. Does not include facilities for processing LPG in non-condensible OH gases (see #14).*
2. Vacuum Crude Distillation	
3. Naphtha Hydrotreating	*For sulfur reduction in straight-run or cracked naphthas.*
4. Middle Distillate Hydrotreating	*For sulfur reduction in jet fuels and kerosene.*
5. Gas Oil Hydrotreating	*For low sulfur fuel oils, cracking feed pretreatment, and lube oil hydroprocessing.*
6. Vacuum Resid Hydrodesulfurization	
7. Catalytic Reforming	*Includes Platforming, Rheniforming, and Powerforming. Does not include naphtha hydrotreating (see #3).*
8. Aromatics Extraction	*Includes Udex, Sulfolane, and Tetra.*
9. Catalytic Cracking	*Includes fluid and moving bed crackers such as the FCC, HCC, and TCC. Includes reactor, regenerator, main fractionater, and heat exchange. Light ends recovery and fractionation are not included (see #14).*
10. Hydrocracking	
11. Thermal Cracking & Visbreaking	
12. Delayed Coking	
13. Fluid Coking	*Includes fluid coking and flexicoking.*
14. Light Ends Recovery and Fractionation	*Includes circulating oil absorption/stripping and fractionation of recovered light ends.*
15. Other Miscellaneous Fractionation Units	*Independent naphtha splitters, rerun stills, stabilizers, etc.*
16. Alkylation	*Includes both HF and H_2SO_4 alkylation.*
17. Polymerization	*Production of polymer gasoline from propylene and LPG mixtures.*
18. Isomerization	*Includes both C_4 and C_5/C_6 isomerization.*
19. Lubes Processing - Volatile Organic Solvents	*Includes propane deasphalting, propane deresining, propane dewaxing, solvent dewaxing, Duo Sol, solvent deasphalting, MEK dewaxing, and MEK-toluene dewaxing.*
20. Other Lube Oil Processing	*Includes phenol extraction, furfural extraction, acid treating, SO_2 extraction, white oil manufacture, centrifuge and chilling, naphthenic lube oils, clay contacting, wax deoiling, wax sweating, wax neutral separation, and compounding.*
21. Asphalt Production	*Includes asphalt oxidizing, asphalt emulsifying, Dubbs pitch, and 200°F softening point unfluxed asphalt.*
22. Hydrogen Production	*Includes steam reforming and partial oxidation.*

Table 6. Refinery Modules (Cont)

Module Name	Comments
23. Gasoline Treating	Includes Merox, inhibitor sweetening, mercapfining, Petreco Locap, Linde, caustic treating, and Doctor treating.
24. Other Product Treating	Includes clay treating, Linde, salt treating, and blending for middle distillates and fuel oils.
25. Olefins Production	Production of mixed olefins from gas, naphtha, and/or oil feedstocks.
26. Other Volatile Petrochemicals	Includes butadiene, alpha olefins, aromatics, cumene, cyclohexane, aliphatics, linear paraffins, heptene, MEK, MIBK, ethyl amyl ketone, tertiary amylenes, acetone, isobutylene, hydrodealkylation of aromatics.
27. Other Low Volatility Petrochemicals	Includes naphthalene, xylenes, mineral spirits, octyl formal alkylate, styrene, phthalic anhydride, nonene, diallylamine, polyisobutylene chloride, oxalcohol, phenol, cresylic acid, naphthenic acid, butyl alcohols, pentoxone, sodium sulfonates, tertiary butyl toluene, polymers, carbon black, furfural, catalysts, mesityl oxide, isophorone, gasoline additives, lubricant additives, and oxidates.
28. Boilers	Independent combustion units for production of steam and/or electricity.
29. Blowdown System and Flares	
30. Wastewater Treating	Includes oil/water separators (OWS) and dissolved air flotation (DAF) units.
31. Sludge/Solids Handling	Includes any on-site treatment such as landfarming, landfilling, and ponding.
32. Crude and Product Storage	Includes fixed roof and floating roof tanks.
33. Cooling Towers	
34. Product Loading Operations	Includes loading facilities for tank trucks, tank cars, barges, ocean barges, and marine tankers.

ods to estimate the accuracy of emission estimates generated by the model.

Obviously, it would be desirable to update the modules periodically as additional emission data become available. Additional emission data from synfuel facilities should be available during the next 5 years. As the model is employed, users will undoubtedly find additional needs which have not been addressed by or included in the current model. These needs should be catalogued for future model improvement efforts.

This VOC emissions model has been evaluated in a preliminary fashion by applying the model to some specific facilities and comparing the emissions estimates to results obtained independently (e.g., through permit procedures). Field tests would be more thorough and objective. Emissions could be estimated using the model and then measured using transect techniques. The results from this effort could be used to refine, calibrate, and validate the model.

Table 7. Synfuel Modules

Module Name	Comments
Coal Preparation (Thermal Drying)	
Slurry Drying	Used in EDS process.
Coal Gasification	Includes gas cooling. Fugitive emissions from some gasifiers negligible because they do not provide significant hydrocarbons.
Methanol Synthesis	
Fischer-Tropsch Synthesis	
Mobil M-Gasoline Synthesis	
Direct Liquefaction	Includes product separation.
Above-Ground Oil-Shale Retorting	
Acid Gas Removal	
Oil-Soluble Arsenic Removal	No default values developed due to lack of process information.
Wastewater Solvent Extraction	Example: Phenosolvan process

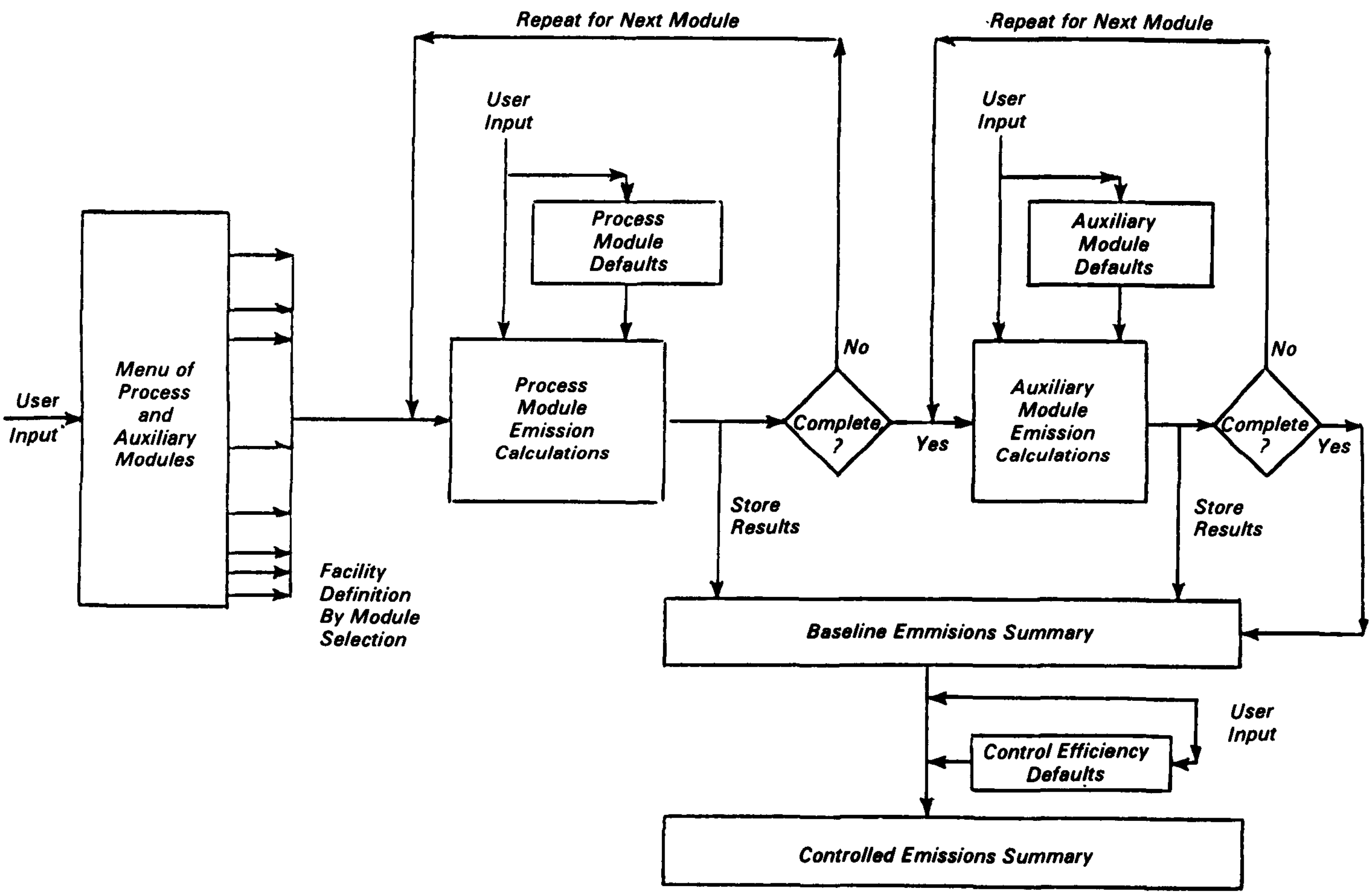

Figure 1. Logic flow diagram.

Table 8. *Modules of Example Small Existing Refinery*[a]

Process Modules:
Atmospheric Crude Distillation
Vacuum Crude Distillation
Naphtha Hydrotreating
Catalytic Reforming
Aromatics Extraction
Fluid Catalytic Cracking
Light Ends Recovery and Fractionation
Other Miscellaneous Fractionation
Alkylation
Auxiliary Modules:
Boiler
Blowdown System and Flares
Wastewater Collection and Treating
Storage--Fixed Roof Tanks
Storage--Floating Roof Tanks
Cooling Towers
Loading Racks--Trucks or Rail Cars

[a]Crude capacity = 50,000 bbl/day.

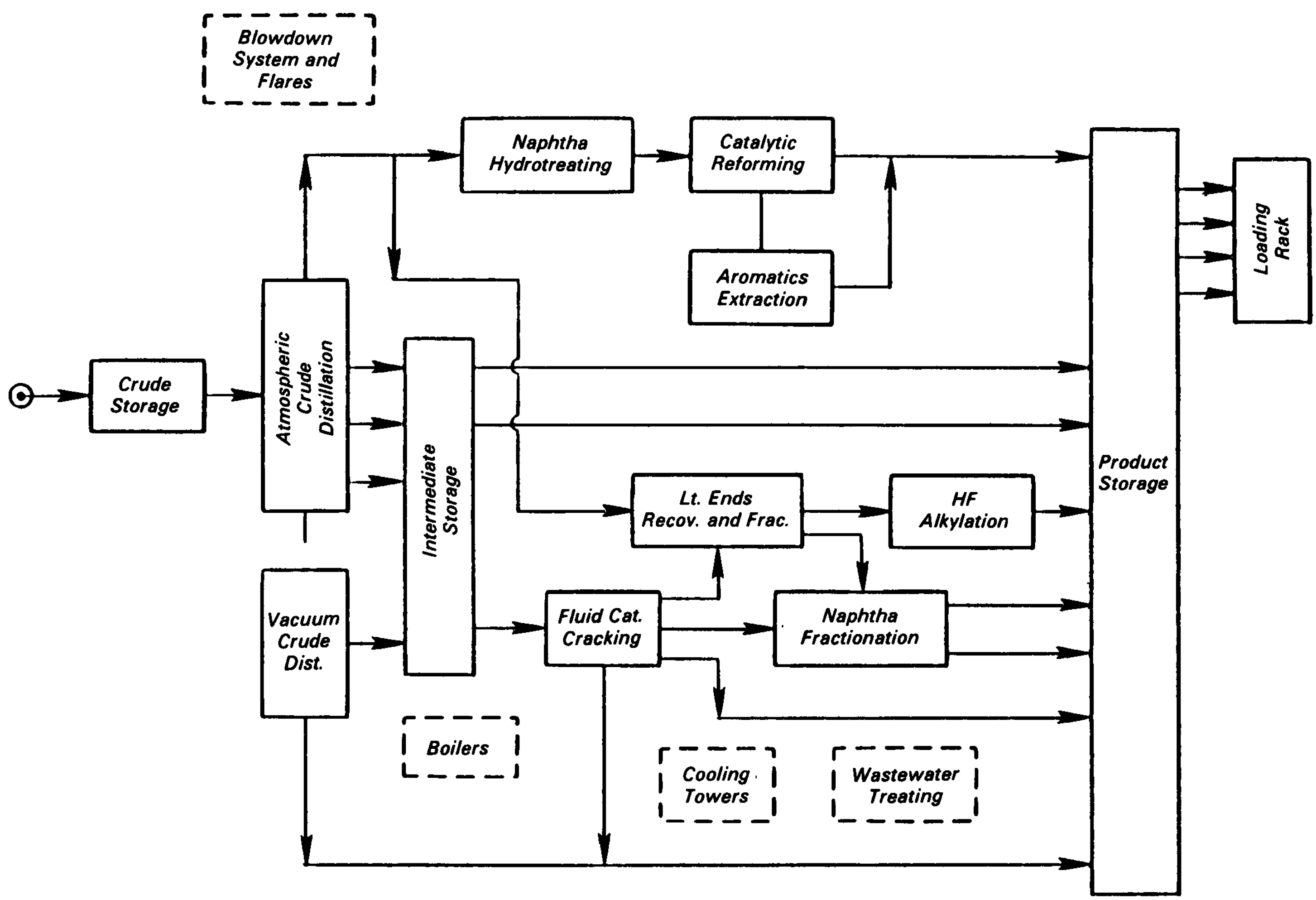

Figure 2. *Block flow diagram for the example small refinery.*

Table 9. *Summary of Baseline Emissions*

Source Type/Service	Emissions, lb/day	Percent of Total
Pumps/Light Liquid	522	2.5
Pumps/Heavy Liquid	53	0.3
Compressors/Hydrocarbon Gas	238	1.1
Compressors/Hydrogen Gas	6	neg.
Valves/Hydrocarbon Gas	4938	23.7
Valves/Hydrogen Gas	142	0.7
Valves/Light Liquids	4950	23.8
Valves/Heavy Liquids	60	0.3
Connections/All	748	3.6
Relief Valves/Gas	1446	6.9
Relief Valves/Liquid	33	0.1
Open-End Lines/All	42	0.2
Process Drains/All	539	2.6
Combustion Sources	91	0.4
Other Point Sources	450	2.2
Wastewater Collection and Treating	1056	5.1
Cooling Towers	1314	6.3
Blowdown System and Flares	110	0.5
Loading Racks	308	1.5
Fixed Roof Storage	1485	7.1
Floating Roof Storage	2306	11.1
Totals	20,837	100.0

Table 10. *Summary of "Maximum Default" Emission Estimates*

The Type A (or topping) refinery can be estimated by:
Emissions (lb/day) = 4,024 + (82.3) (Crude Rate in 10^3 BPD[a])
The average Type A refinery has a crude capacity of 14,000 BPSD[b].

The Type B (or cracking) refinery can be estimated by:
Emissions (lb/day) = 13,649 + (82.4) (Crude Rate in 10^3 BPD)
The average Type B refinery has a crude capacity of 66,000 BPSD.

The Type C (or petrochemicals) refinery can be estimated by:
Emissions (lb/day) = 25,339 + (83.1) (Crude Rate in 10^3 BPD)
The average Type C refinery has a crude capacity of 150,000 BPSD.

The Type D (or lubes) refinery can be estimated by:
Emissions (lb/day) = 24,455 + (86.0) (Crude Rate in 10^3 BPD)
The average Type D refinery has a crude capacity of 187,000 BPSD.

The Type E (or integrated) refinery emissions can be estimated by:
Emissions (lb/day) = 30,114 + (86.5) (Crude Rate in 10^3 BPD)
The average Type E refinery has a crude capacity of 312,000 BPSD.

[a]Barrels per day.
[b]Barrels per stream day..

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The complete report consists of two volumes, entitled "A Model for Evaluation of Refinery and Synfuels VOC Emissions Data:"
"Volume I. Technical Report and Appendix A," (Order No. PB 85-215 713/AS; Cost: $23.50)
"Volume II. Appendices B and C," (Order No. PB 85-215 721/AS; Cost: $16.00)
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