[NOTE: With the publication of the Fifth Edition of AP-42, the Chapter and Section number for Synthetic Ammonia changed to 8.1]

BACKGROUND REPORT

AP-42 SECTION 5.2

SYNTHETIC AMMONIA

Prepared for

U.S. Environmental Protection Agency OAQPS/TSD/EIB Research Triangle Park, NC 27711

1-96

Pacific Environmental Services, Inc. P.O. Box 12077 Research Triangle Park, NC 27709 919/941-0333

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AP-42 Background Report

TECHNICAL SUPPORT DIVISION

U.S. ENVIRONMENTAL PROTECTION AGENCY Office of Air Quality Planning and Standards Research Triangle Park, NC 27711 This report has been reviewed by the Technical Support Division of the Office of Air Quality Planning and Standards, EPA. Mention of trade names or commercial products is not intended to constitute endorsement or recommendation for use. Copies of this report are available through the Library Services Office (MD-35), U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

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1.0 INTRODUCTION

The document "Compilation of Air Pollutant Emission Factors" (AP-42) has been published by the U.S. Environmental Protection Agency (EPA) since 1972. Supplements to AP-42 have been routinely published to add new emission source categories and to update existing emission factors. AP-42 is routinely updated by the EPA to respond to new emission factor needs of the EPA, State and local air pollution control programs, and industry.

An emission factor relates the quantity (weight) of pollutants emitted to a unit of activity of the source. The uses for the emission factors reported in AP-42 include:

- 1. Estimates of area-wide emissions;
- 2. Emission estimates for a specific facility; and
- 3. Evaluation of emissions relative to ambient air quality.

The purpose of this report is to provide background information from process information obtained from industry comment to support revision of emission factors for synthetic ammonia.

Including the introduction (Chapter 1), this report contains four chapters. Chapter 2 gives a description of the synthetic ammonia industry. It includes a characterization of the industry, an overview of the different process types, a description of emissions, and a description of the technology used to control emissions resulting from synthetic ammonia.

Chapter 3 is a review of emissions data collection and analysis procedures. It describes the literature search, the screening of emission data reports, and the quality rating system for both emission data and emission factors. Chapter 4 details criteria and noncriteria pollutant emission factor development. It includes the review of specific data sets and the results of data analysis. Particle size determination and particle size data analysis methodology are described when applicable. Appendix A presents AP-42 Section 5.2.

2.0 INDUSTRY DESCRIPTION

2.1 GENERAL^{1,2}

Synthetic ammonia (NH₃) refers to ammonia that has been synthesized (SIC 2873) from natural gas. Natural gas molecules are reduced to carbon and hydrogen. The hydrogen is then purified and reacted with nitrogen to produce ammonia. Approximately 75 percent of the ammonia produced is used as fertilizer, either directly as ammonia or indirectly after synthesis as urea, ammonium nitrate, and monoammonium or diammonium phosphates. The remaining is used as raw material in the manufacture of polymeric resins, explosives, nitric acid, and other products.

Synthetic ammonia plants are located throughout the U.S. and Canada. Synthetic ammonia is produced in 25 states by 60 plants which have an estimated combined annual production capacity of 15.9 million megagrams (17.5 million tons) in 1991. Ammonia plants are concentrated in areas with abundant supplies of natural gas. Seventy percent of U.S. capacity is located in Louisiana, Texas, Oklahoma, Iowa and Nebraska. Table 2.2-1 shows the distribution of synthetic ammonia plants, along with 1991 capacity.²

2.2 PROCESS DESCRIPTION^{1,3,4}

Anhydrous ammonia is synthesized by reacting hydrogen with nitrogen at a molar ratio of 3 to 1, then compressing the gas and cooling it to -33 °C (-27 °F).³ Nitrogen is obtained from the air, while hydrogen is obtained from either the catalytic steam reforming of natural gas (methane) or naphtha, or the electrolysis of brine at chlorine plants. In the U.S., about 98 percent of synthetic ammonia is produced by catalytic steam reforming of natural gas.¹ Figure 2.2-1 shows a general process flow diagram of a typical ammonia plant.³

Six process steps are required to produce synthetic ammonia using the catalytic steam reforming method: 1) natural gas desulfurization, 2) catalytic steam reforming, 3) carbon monoxide shift, 4) carbon dioxide removal, 5) methanation and 6) ammonia synthesis.³ The first, third, fourth, and fifth steps remove impurities such as sulfur, CO, CO_2 and water from the feedstock, hydrogen and synthesis gas streams. In the second step, hydrogen is manufactured and nitrogen (air) is introduced into this two stage process.

Facility name/location	Total capacity (10 ³ tons/yr)
Unocal Corp., Kenai, AK	1200
LaRoche Industries, Inc., Cherokee, AL	175
Agrico Chemical, Blytheville, AR	b
Agricultural Minerals Corp., Blytheville, AR	400
Nitrogen Products, Inc., Helena, AR	210
Chevron Chemical Co., El Segundo, CA	20
Air Products & Chemical, Inc., Pace Junction, FL	100
Arcadian Corp., Augusta, GA	545
Columbia Nitrogen Corp., Augusta, GA	b
Arcadian Corp., Clinton, IA	260
Farmland Industries, Inc., Fort Dodge, IA	210
Green Valley Chem Corp., Creston, IA	35
Hawkeye Chemical Co., Clinton, IA	b
Terra International, Inc., Port Neal, IA	230
Simplot, J.R., Co., Pocatello, ID	108
Coal gasification, Girard, IL	b
Phoenix Chemical Co., East Dubuque, IL	238
Farmland Industries, Inc., Dodge City, KS	210
Farmland Industries, Inc., Lawrence, KS	350
Agrico Chemical, Donaldsonville, LA	450
Air Products & Chemical, Inc., New Orleans, LA	290
American Cyanamid Co., Fortier, LA	425
Arcadian Corp., Geismar, LA	410
Arcadian Corp., Lake Charles, LA	440
Borden Chemicals & Plastics, Geismar, LA	400
CF Industries, Inc., Donaldsonville, LA	1770
Farmland Industries, Inc., Pollock, LA	420
First Mississippi Fertilizer Co., Donaldsonville, LA	450
IMC Fertilizer, Inc., Sterlington, LA	1050
Jupiter Chemicals, Inc., Westlake, LA	30
Monsanto Co., Luling, LA	490

 TABLE 2.1-1

 DISTRIBUTION OF SYNTHETIC AMMONIA PLANTS WITH 1992 CAPACITY^a

^a Reference 2.

^b These plants have been sold and production capacity data for 1991 are not available.

Facility name/location	Total capacity (10 ³ tons/yr)
Olin Corp., Lake Charles, LA	b
Triad Chemical Co., Donaldsonville, LA	450
Mississippi Chemical Corp., Yazoo City, MS	500
Ang Coal Gas, Beulah, ND	24
Arcadian Corp., Laplatte, NE	190
Cepex, Inc., Beatrice, NE	b
Farmland Industries, Inc, Beatrice, NE	237
BP Chemical, Lima, OH	530
Agrico Chemical, Verdigris, OK	b
Agricultural Minerals Corp., Verdigris, OK	1000
Farmland Industries, Inc., Enid, OK	840
Grace-Oklahoma Nitrogen, Woodward, OK	b
Terra International, Inc., Woodward, OK	450
Wil-Grow Fertilizer Co., Pryor, OK	94
Atochem North America, Portland, OR	8
Chevron Chemical Co., St. Helens, OR	80
Carbonaire Co., Inc, Palmerton, PA	35
Arcadian Corp., Woodstock, TN	340
Grace, W.R., & Co., Woodstock, TN	b
Nitrex, Woodstock, TN	b
Center Plains Industries, Dumas, TX	b
Cominco, Borger, TX	400
Diamond Shamrock, Dumas, TX	b
DuPont & Co., Inc., Beaumont, TX	490
Allied-Signal Corp., Hopewell, VA	410
Chevron Chemical Co., Finley, WA	140
Oxychem Electrochemicals, Tacoma, WA	28
PPG Industries, Inc., Natrium, WV	50
Coastal Chem, Inc., Cheyenne, WY	170

TABLE 2.1-1 (concluded)DISTRIBUTION OF SYNTHETIC AMMONIA PLANTS WITH 1992 CAPACITY^a

^aReference 2.

^bThese plants have been sold and production capacity data for 1991 are not available.

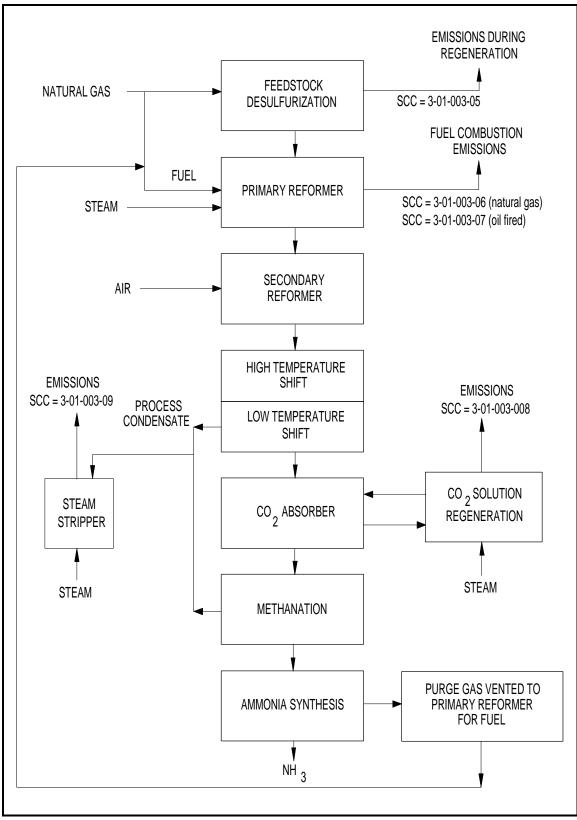


Figure 2.2-1 General process flow of a typical ammonia plant.

The sixth step produces anhydrous ammonia from the synthetic gas. While all ammonia plants use this basic process, details such as operating pressures, temperatures, and quantities of feedstock vary from plant to plant.

Natural gas desulfurization

In this step, the sulfur content (as H₂S) in natural gas is reduced to below 280 micrograms per cubic meter to prevent poisoning of the nickel catalyst in the primary reformer.¹ Desulfurization (SCC 3-01-003-05) can be accomplished by using either activated carbon or zinc oxide. Over 95 percent of the ammonia plants in the U.S. use activated carbon fortified with metallic oxide additives for feedstock desulfurization.³ The remaining plants use a tank filled with zinc oxide for desulfurization.³ Heavy hydrocarbons can decrease the effectiveness of an activated carbon bed. This carbon bed also has another disadvantage in that it cannot remove carbonylsulfide. Regeneration of carbon is accomplished by passing superheated steam through the carbon bed. A zinc oxide bed offers several advantages over the activated carbon bed. Steam regeneration to use as energy is not required when using a zinc oxide bed. No air emissions are created by the zinc oxide bed, and the higher molecular weight hydrocarbons are not removed; therefore, the heating value of the natural gas is not reduced.

Catalytic steam reforming

Natural gas leaving the desulfurization tank is mixed with process steam and preheated to 540° C (1004° F). The mixture of steam and gas enters the primary reformer (natural gas fired primary reformer SCC 3-01-003-06 and oil fired primary reformer SCC 3-01-003-07) tubes, which are filled with a nickel-based reforming catalyst. Approximately 70 percent of the methane (CH₄) is converted to hydrogen and carbon dioxide (CO₂).³ An additional amount of CH₄ is converted to CO. This process gas is then sent to the secondary reformer, where it is mixed with compressed air that has been preheated to about 540° C (1004° F). Sufficient air is added to produce a final synthesis gas having a hydrogen-to-nitrogen mole ratio of 3 to 1. The gas leaving the secondary reformer is then cooled to 360° C (680° F) in a waste heat boiler.³

Carbon monoxide shift

After cooling, the secondary reformer effluent gas enters a high temperature CO shift converter which is filled with chromium oxide initiator and iron oxide catalyst. The following reaction takes place in the carbon monoxide converter:

$$CO + H_2O \rightarrow CO_2 + H_2 \tag{1}$$

The exit gas is then cooled in a heat exchanger. In some plants, the gas is passed through a bed of zinc oxide to remove any residual sulfur contaminants that would poison the low temperature shift catalyst. In other plants, excess low temperature shift catalyst is added to ensure that the unit will operate as expected. The low temperature shift converter is filled with a copper oxide/zinc oxide catalyst. Final shift gas from this converter is cooled from 210 to 110° C (410 to 230° F) and enters the bottom of the carbon dioxide absorption system. Unreacted steam is condensed and separated from the gas in a knockout drum. This condensed steam (process condensate) contains ammonium carbonate ([(NH₄)₂ CO₃ • H₂O]) from the high temperature shift converter, methanol (CH₃OH) from the low temperature shift converter, and small amounts of sodium, iron, copper, zinc, aluminum and calcium.

Process condensate is sent to the stripper (SCC 3-01-003-09) to remove volatile gases such as ammonia, methanol, and carbon dioxide. Trace metals remaining in the process condensate are removed by the ion exchange unit.

Carbon dioxide removal

In this step, CO_2 in the final shift gas is removed. CO_2 removal can be done by using two methods: monoethanolamine ($C_2H_4NH_2OH$) scrubbing and hot potassium scrubbing. Approximately 80 percent of the ammonia plants use monoethanolamine (MEA) to aid in removing CO_2 .³ The CO_2 gas is passed upward through an adsorption tower countercurrent to a 15 percent to 30 percent solution of MEA in water fortified with effective corrosion inhibitors. After absorbing the CO_2 , the amine solution is preheated and regenerated (carbon dioxide regenerator SCC 3-01-003-08) in a reactivating tower. This reacting tower removes CO_2 by steam stripping and then by heating. The CO_2 gas (98.5 percent CO_2) is either vented to the atmosphere or used for chemical feedstock in other parts of the plant complex. The regenerated MEA is pumped back to the absorber tower after being cooled in a heat exchanger and solution cooler.³

Methanation

Residual CO_2 in the synthesis gas is removed by catalytic methanation which is conducted over a nickel catalyst at temperatures of 400 to 600°C (752 to 1112°F) and pressures up to 3,000 kPa (435 psia) according to the following reactions:

$$CO + 3H_2 \rightarrow CH_4 + H_2O \tag{2}$$

$$CO_2 + H_2 \rightarrow CO + H_2O \tag{3}$$

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \tag{4}$$

Exit gas from the methanator, which has a 3:1 mole ratio of hydrogen and nitrogen, is then cooled to $38^{\circ}C$ (100°F).

Ammonia Synthesis

In the synthesis step, the synthesis gas from the methanator is compressed at pressures ranging from 13,800 to 34,500 kPa (2000 to 5000 psia), mixed with recycled synthesis gas, and cooled to 0°C (32°F). Condensed ammonia is separated from the unconverted synthesis gas in a liquid-vapor separator and sent to a let-down separator. The unconverted synthesis is compressed and preheated to 180°C (356°F) before entering the synthesis converter which contains iron oxide catalyst. Ammonia from the exit gas is condensed and separated, then sent to the let-down separator. A small portion of the overhead gas is purged to prevent the buildup of inert gases such as argon in the circulating gas system.

Ammonia in the let-down separator is flashed to 100 kPa (14.5 psia) at -33 °C (-27 °F) to remove impurities from the liquid. The flash vapor is condensed in the let-down chiller where anhydrous ammonia is drawn off and stored at low temperature.

2.3 EMISSIONS AND CONTROLS^{1,3}

Pollutants from the manufacture of synthetic anhydrous ammonia are emitted from four process steps: 1) regeneration of the desulfurization bed, 2) heating of the catalytic steam, 3) regeneration of carbon dioxide scrubbing solution, and 4) steam stripping of process condensate.

More than 95 percent of the ammonia plants in the U.S. use activated carbon fortified with metallic oxide additives for feedstock desulfurization.³ The desulfurization bed must be regenerated about once every 30 days for an average period of 8 to 10 hours. Vented regeneration steam contains sulfur oxides (SO_x) and hydrogen sulfide (H₂S), depending on the amount of oxygen in the steam. Regeneration also emits hydrocarbons and carbon monoxide (CO). The reformer, heated with natural gas or fuel oil, emits combustion products such as NO_x, CO, SO_x, hydrocarbons, and particulates.

Carbon dioxide (CO₂) is removed from the synthesis gas by scrubbing with monoethanolamine ($C_2H_4NH_2OH$) or hot potassium carbonate solution. Regeneration of this CO₂ scrubbing solution with steam produces emission of water, NH₃, CO, CO₂ and monoethanolamine.

Cooling the synthesis gas after low temperature shift conversion forms a condensate containing NH_3 , CO_2 , methanol (CH_3OH), and trace metals. Condensate steam strippers are used to remove NH_3 and methanol from the water, and steam from this is vented to the atmosphere, emitting NH_3 , CO_2 , and methanol.

Some processes have been modified to reduce emissions and to improve utility of raw materials and energy. One such technique is the injection of the overheads into the reformer stack along with the combustion gases to eliminate emissions from the condensate steam stripper.

2.4 **REVIEW OF SPECIFIC REFERENCES**

Pacific Environmental Services (PES) contacted the following sources to obtain the most up-to-date information on process descriptions and emissions for this industry:

- 1) Alabama Air Division, Montgomery, Alabama.
- 2) Allied-Signal, Hopewell, Virginia.
- 3) Arkansas Department of Air Pollution Control and Ecology.
- 4) Chemical Manufacturers Association.
- 5) Florida Department of Environmental Regulation, Tallahassee, Florida.
- 6) Georgia Department of Natural Resources, Atlanta, Georgia.

- 7) J.R. Simplot, Co., Pocatello, Idaho.
- 8) Kansas Department of Health and Environment, Topeka, Kansas.
- 9) Michigan Department of Natural Resources, Lansing, Michigan.
- 10) Missouri Department of Natural Resources, Jefferson City, Missouri.
- 11) Ohio Environmental Protection Agency, Ohio.
- 12) Pennsylvania Department of Environmental Resources, Harrisburg, Pennsylvania.
- 13) BP Chemicals, Lima, Ohio.

Responses were received from the Ohio Environmental Protection Agency and BP Chemicals. No responses were received from the remaining sources. PES did not incorporate the information received from these sources into the AP-42 chapter revision. The Ohio Environmental Protection Agency provided one source test which did not contain enough material (no documentation of EPA Methods 1 through 4 or testing method, no field data, etc.) to use as a valid test. BP Chemicals also provided a source test, but it did not contain enough material (no documentation of EPA Methods 1 through 4, no field data, no calibration data, no equipment prep documentation, and no process description) to use as a valid source test.

The following references were used to establish process descriptions, emissions, controls, and a general characterization of the synthetic ammonia industry. A brief discussion follows each reference. These discussions provide a summary of the information gathered as background information for the AP-42 Section 5.2.

Reference 1: Source Category Survey: Ammonia Manufacturing Industry

This reference provided a generalized description of the synthetic ammonia process. In the U.S., about 98 percent of synthetic ammonia is produced by catalytic steam reforming of natural gas. Natural gas is converted to hydrogen, purified, and reacted with nitrogen to produce ammonia. Approximately 75 percent of ammonia produced is used as fertilizer, either directly as ammonia or indirectly after synthesis as urea, ammonium nitrate and monoammonium or diammonium phosphates. The remaining is used as raw material in the manufacture of polymeric resins, explosives, nitric acid and other products. In the desulfurization step, the sulfur content (as H_2S) in natural gas is reduced to below 280 micrograms per cubic meter to prevent poisoning the nickel catalyst in the primary reformer. The carbon bed which is often used in this step has several disadvantages: heavy hydrocarbons can decrease the effectiveness of an activated carbon bed and

the bed cannot remove carbonylsulfide. When zinc beds are used instead of carbon, there are several advantages. Steam regeneration is not necessary. No air emissions are created by the zinc oxide bed, and the higher molecular weight hydrocarbons are not removed; therefore, the heating value of the natural gas is not reduced.

Reference 2: North American Fertilizer Capacity Data

This reference provided information on quantities of synthetic ammonia plants, their location, and their production capacity. There are 60 synthetic ammonia plants located throughout the U.S. and Canada. However, some of these plants have been sold, and production capacity information is unavailable. Seventy percent of the U.S. capacity is located in Louisiana, Texas, Oklahoma, Iowa, and California. The total annual U.S. production of ammonia in 1991 is estimated to be 15.9 million megagrams (17.5 million tons), based on capacities of plants which have not been sold. This information has been incorporated into the revised 1992 AP-42.

Reference 3: Source Assessment: Synthetic Ammonia Production

This reference provided the majority of the process description and emissions details. Over 95 percent of the ammonia plants in the U.S. use activated carbon fortified with metallic oxide additives for feedstock desulfurization. The remaining plants use a tank filled with zinc oxide for desulfurization. Approximately 70 percent of the methane (CH_4) is converted to hydrogen and carbon dioxide (CO_2) . The remaining discussion for catalytic steam reforming, carbon monoxide shift, carbon dioxide removal, methanation, and ammonia synthesis came from this reference. The process steps from which pollutants may be emitted are also detailed in this reference. Some processes have been modified to reduce emissions and to improve utility of raw materials and energy. One such technique is the injection of the overheads into the reformer stack along with the combustion gases to eliminate emissions from the condensate steam stripper.

<u>Reference 4</u>: <u>AIRS Facility Subsystem Source Classification Codes and Emission Factor Listing</u> For Criteria Pollutants.

AIRS provided SIC and SCC codes for the synthetic ammonia manufacturing process. Some of the basic steps of the manufacturing process are not listed in AIRS. The secondary reformer, high temperature CO_2 shift converter and heat exchanger, low temperature CO_2 shift converter, CO_2 absorber (monoethanolamine scrubber and hot potassium scrubber), methanation step (nickel catalyst at high temperature and pressure), ammonia synthesis (compression of synthesis gas at extremely high pressure and temperature of 0°C), and its let-down liquid-vapor separator were not listed with an SCC code.

2.5 **REFERENCES FOR CHAPTER 2**

- 1. <u>Source Category Survey: Ammonia Manufacturing Industry</u>, EPA-450/3-80-014, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, August 1980.
- 2. <u>North American Fertilizer Capacity Data</u>, Tennessee Valley Authority, Muscle Shoals, Alabama, December 1991.
- G.D. Rawlings and R.B. Reznik, <u>Source Assessment: Synthetic Ammonia Production</u>, EPA-600/2-77-107m, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, November 1977.
- 4. <u>AIRS Facility Subsystem Source Classification Codes and Emission Factor Listing For</u> <u>Criteria Pollutants</u>. EPA-450/4-90-003, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, March 1990.

3.0 GENERAL EMISSION DATA REVIEW AND ANALYSIS PROCEDURES

3.1 LITERATURE SEARCH AND SCREENING

The first step of this investigation involved a search of available literature relating to criteria and noncriteria pollutant emissions associated with synthetic ammonia. This search included the following references:

- 1) AP-42 background files maintained by the Emission Factor and Methodologies Section.
- 2) Files maintained by the Emission Standards Division.
- 3) Information in the *Air Facility Subsystems* (AFS) of the EPA *Aerometric Information Retrieval System* (AIRS).
- 4) References in the National Technical Information Service (NTIS).
- 5) Handbook of Emission Factors, Parts I and II, Ministry of Housing, Physical Planning, and Environment, The Netherlands, 1980/1983.

To reduce the amount of literature collected to a final group of references pertinent to this report, the following general criteria were used:

- Emissions data must be from a primary reference, i.e. the document must constitute the original source of test data. For example, a technical paper was not included if the original study was contained in the previous document.
- 2. The referenced study must contain test results based on more than one test run.
- 3. The report must contain sufficient data to evaluate the testing procedures and source operating conditions (e.g., one-page reports were generally rejected).

If no primary data was found and the previous update utilized secondary data, this secondary data was still used and the Emission Factor Rating lowered, if needed. A final set of reference materials was compiled after a thorough review of the pertinent reports, documents, and information according to these criteria. The final set of reference materials is given in Chapter 4.5.

3.2 EMISSION DATA QUALITY RATING SYSTEM

As part of Pacific Environmental Services' analysis of the emission data, the quantity and quality of the information contained in the final set of reference documents were evaluated. The following data were always excluded from consideration.

- Test series averages reported in units that cannot be converted to the selected reporting units;
- 2. Test series representing incompatible test methods (i.e., comparison of the EPA Method 5 front-half with the EPA Method 5 front- and back-half);
- 3. Test series of controlled emissions for which the control device is not specified;
- 4. Test series in which the source process is not clearly identified and described; and
- 5. Test series in which it is not clear whether the emissions were measured before or after the control device.

Data sets that were not excluded were assigned a quality rating. The rating system used was that specified by the OAQPS for the preparation of AP-42 sections. The data were rated as follows:

A

Multiple tests performed on the same source using sound methodology and reported in enough detail for adequate validation. These tests do not necessarily conform to the methodology specified in either the inhalable particulate (IP) protocol documents or the EPA reference test methods, although these documents and methods were certainly used as a guide for the methodology actually used.

B

Tests that were performed by a generally sound methodology but lack enough detail for adequate validation.

С

Tests that were based on an untested or new methodology or that lacked a significant amount of background data.

D

Tests that were based on a generally unacceptable method but may provide an order-ofmagnitude value for the source. The following criteria were used to evaluate source test reports for sound methodology and adequate detail:

- 1. <u>Source operation</u>. The manner in which the source was operated is well documented In the report. The source was operating within typical parameters during the test.
- 2. <u>Sampling procedures</u>. The sampling procedures conformed to a generally acceptable methodology. If actual procedures deviated from accepted methods, the deviations are well documented. When this occurred, an evaluation was made of the extent such alternative procedures could influence the test results.
- 3. <u>Sampling and process data</u>. Adequate sampling and process data are documented in the report. Many variations can occur unnoticed and without warning during testing. Such variations can induce wide deviations in sampling results. If a large spread between test results cannot be explained by information contained in the test report, the data are suspect and were given a lower rating.
- 4. <u>Analysis and calculations</u>. The test reports contain original raw data sheets. The nomenclature and equations used were compared to those (if any) specified by the EPA to establish equivalency. The depth of review of the calculations was dictated by the reviewer's confidence in the ability and conscientiousness of the tester, which in turn was based on factors such as consistency of results and completeness of other areas of the test report.

3.3 EMISSION FACTOR QUALITY RATING SYSTEM

The quality of the emission factors developed from analysis of the test data was rated utilizing the following general criteria:

A (Excellent)

Developed only from A-rated test data taken from many randomly chosen facilities in the industry population. The source category is specific enough so that variability within the source category population may be minimized.

B (Above average)

Developed only from A-rated test data from a reasonable number of facilities. Although no specific bias is evident, it is not clear if the facilities tested represent a random sample of

the industries. As in the A-rating, the source category is specific enough so that variability within the source category population may be minimized.

C (Average)

Developed only from A- and B-rated test data from a reasonable number of facilities. Although no specific bias is evident, it is not clear if the facilities tested represent a random sample of the industry. As in the A-rating, the source category is specific enough so that variability within the source category population may be minimized.

D (Below average)

The emission factor was developed only from A- and B-rated test data from a small number of facilities, and there is reason to suspect that these facilities do not represent a random sample of the industry. There also may be evidence of variability within the source category population. Limitations on the use of the emission factor are noted in the emission factor table.

E (Poor)

The emission factor was developed from C- and D-rated test data, and there is reason to suspect that the facilities tested do not represent a random sample of the industry. There also may be evidence of variability within the source category population. Limitations on the use of these factors are always noted.

The use of these criteria is somewhat subjective and depends to an extent on the individual reviewer.

3.4 **REFERENCES FOR CHAPTER 3**

- <u>Technical Procedures for Developing AP-42 Emission Factors and Preparing AP-42</u> <u>Sections</u>. U.S. Environmental Protection Agency, Emissions Inventory Branch, Office of Air Quality Planning and Standards, Research Triangle Park, NC 27711, April 1992. [Note: this document is currently being revised at the time of this printing.]
- 2. <u>AP-42</u>, Supplement A, Appendix C.2, "Generalized Particle Size Distributions." U.S. Environmental Protection Agency, October 1986.

4.0 POLLUTANT EMISSION FACTOR DEVELOPMENT

4.1 **REVIEW OF SPECIFIC DATA SETS**

Two source tests were received for review to update the present AP-42 section. One was provided by the Ohio Environmental Protection Agency, and the other by BP Chemicals. Neither of these tests provided enough detail to use as valid reports as dictated by the general criteria presented in Chapter 3 of this report.

Reference 1: Source Category Survey: Ammonia Manufacturing Industry

This reference is listed as Reference 2 in the 1983 AP-42 and was used to develop its emissions table. It provides emission factors for desulfurization, the reformer (PES assumes primary), the steam stripper (condensate stripper), and the CO_2 absorber in the synthetic ammonia manufacturing process. These emissions are based on Reference 2 of this section and a study of seven plants.

Desulfurization

The desulfurization emission factors are presented for SO_2 , CO, and VOC. An SO_2 emission factor is not given in the 1983 AP-42. This reference states that its SO_2 emission factor originates from another reference (Reference 2 of this section). After review of Reference 2, PES could not verify that the SO_2 emission factor came from this reference. For this reason, this SO_2 emission factor for desulfurization will not be added to the revised 1992 AP-42. The CO emission factor provided by this reference agrees with the factor given in the 1983 AP-42.

The VOC emission factor given in this reference (Reference 1) for desulfurization is 3.6 grams of VOC per kilogram of ammonia produced. Reference 1 states that this factor originates from Reference 2 of this section. After review of Reference 2, PES determined that the factor has been transferred incorrectly into Reference 1. The factor given in Reference 2 is given as hydrocarbons, not VOC. Since neither of these references provide any information on the methane or nonmethane content in the hydrocarbon total of Reference 2, PES has concluded that the 1983 AP-42 has listed the methane and NMOC emission factors incorrectly. The revised 1992 AP-42 will present the hydrocarbon emission factor under total organic compounds (TOC) which may include VOC emissions (or NMOC emissions).

Primary Reformer

The emission factors given for the reformer (PES assumes natural gas primary reformer) are for SO₂, NO_x, CO, VOC, and TSP (total suspended particulate). The emission factors for NO_x and CO are the same as those presented in the 1983 AP-42 and have been changed as described in the discussion under Reference 2. The SO₂ emission factor in this reference matches the 1983 AP-42 SO_x emission factor. Since this is the reference which was used to develop the AP-42 emission factor (along with Reference 2 of this section) and the emission value is given in terms of SO₂ in both references, the revised 1992 AP-42 will footnote this factor to contain mostly SO₂.

The VOC emission factor given in this reference (Reference 1) for the reformer is 0.012 grams of VOC per kilogram of ammonia produced. Reference 1 states that this factor originates from Reference 2 of this section. After review of Reference 2, PES determined that the factor has been transferred incorrectly into Reference 1. The factor given in Reference 2 is given as hydrocarbons, not VOC. Since neither of these references provide any information on the methane or nonmethane content in the hydrocarbon total of Reference 2, PES has concluded that the 1983 AP-42 has listed the methane and NMOC emission factors incorrectly. The revised 1992 AP-42 will present this factor as NMOC under the TOC emission heading. For additional details, see discussion under Reference 2 in this section.

Condensate Stripper

The emissions given for the steam stripper (condensate stripper) are for methanol and ammonia. The methanol emission factor in this reference is presented in the 1983 AP-42 as NMVOC and is footnoted to contain mostly methanol. Since NMVOC is a subset of NMOC, methanol emissions will be presented under the TOC emission heading and footnoted to contain mostly methanol is also classified as a hazardous air pollutant (HAP), and will be footnoted as such in the revised 1992 AP-42. The ammonia emissions in this reference concur with the emission factor presented in the 1983 AP-42; therefore, the revised 1992 AP-42 ammonia emission factor will remain unchanged.

CO₂ Absorber

The emissions given for the CO_2 absorber are for CO, VOC, monoethanolamine, and ammonia. Emission factors for this segment of the synthetic ammonia manufacturing process are

not provided in the 1983 AP-42; instead, emissions are indicated to originate from the CO_2 regenerator. For details on these emissions, see discussion on the CO_2 regenerator for Reference 2 of this section.

Reference 2: Source Assessment: Synthetic Ammonia Production

This reference is Reference 1 in the 1983 AP-42 section for synthetic ammonia and was used to develop the emission factor table in AP-42. Emissions are provided for several stages of the synthetic ammonia manufacturing process: the desulfurization tank, primary reformer (natural gas fired and fuel oil fired), carbon dioxide (CO_2) regenerator, and the condensate stripper.

Desulfurization

The desulfurization process can emit sulfur, carbon monoxide (CO), and hydrocarbons as a result of carbon bed regeneration. The emission factors presented for this process are based on information from an ammonia plant which produces 480 metric tons of ammonia per day (assuming one day equals 24 hours). Sulfur emissions are presented as total sulfur released while assuming a worst case scenario that all the sulfur entering the tank is released during regeneration. This sulfur may include SO_x and H₂S. The emission factor provided by this reference for total sulfur is equivalent to the emission factor presented in the 1983 AP-42. PES does not have the references used to derive this factor; however, from the information contained in this reference (Reference 2), PES could verify that this factor was calculated correctly based on the assumptions listed in the reference. The total sulfur emission factor will remain unchanged in the revised 1992 AP-42, but the emission factor rating will be lowered to "E." This factor rating was lowered since source test data were not used to determine the emissions; instead, emissions were based on secondary references (personal communications, journals, and a handbook of industrial chemistry).

Carbon monoxide and hydrocarbon emissions are based on one set of source test measurements in the public files at the Texas Air Control Board. The CO emission factor is equivalent to the emission factor presented in the 1983 AP-42. Since the source tests used to calculate this factor were not properly referenced, the emission factor rating will be lowered to an "E" in the revised 1992 AP-42. The hydrocarbon emission factor rating will also be lowered to an "E" for the same reason. These hydrocarbon emissions are reported incorrectly in the 1983 AP-42 as nonmethane organic compounds (NMOC). Hydrocarbon emissions in Reference 3 were measured as methane. In other words, total hydrocarbons are reported in terms of methane equivalents; the individual species were not determined. Therefore, the portion of these hydrocarbons that may actually be methane is not known. In the revised 1992 AP-42, these hydrocarbon emissions will be reported as total organic compounds (TOC), and not as NMOC.

Primary Reformer (Natural Gas)

The natural gas fired primary reformer emits SO_x , NO_x , carbon monoxide (CO), nonmethane organic compounds (NMOC), methane (CH₄), and filterable particulate. Emissions in the 1983 AP-42 are based on this reference and the 1973 AP-42 Section 1.4 Natural Gas Combustion. Emission factors in the AP-42 Section 1.4 are calculated based on an average natural gas heating value of 8270 kcal/m³ (1000 Btu/scf) and are given in terms of mass of emissions per volume of fuel fired for a small industrial boiler. This emission factor was then manipulated to reflect emissions in terms of mass of emissions per mass of ammonia produced. Reference 3 provides the combustion rate needed to convert the values given in the AP-42 Section 1.4. The combustion rate is given for a range of 200 to 300 cubic meters of fuel per metric ton of ammonia produced (7.4x10⁶ to 9.8x10⁶ Btu per ton of ammonia produced). To calculate the revised emission factors using the current AP-42 Section 1.4 (revised October 1992) an average combustion rate of 250 cubic meters of fuel per metric ton of ammonia produced was used. This combustion rate is multiplied by the emission factors for small industrial boilers given in the AP-42 Section 1.4 and any applicable conversion factors to yield the final emission factors which are reported in the revised 1992 AP-42 Section 5.2 Synthetic Ammonia.

The SO_x, NO_x, and CO emission factor ratings will remain an "A" as dictated by the emission factor ratings given in the current AP-42 Section 1.4. The NMOC emission factor was calculated based on 48 percent of TOC as stated in Table 1.4-3 and will be placed under the TOC heading. The methane emission factor was calculated based on 52 percent of TOC. Both NMOC and methane emission factors will be given a rating of "C" as dictated by the emission factor was calculated using the upper limit of the emission range specified in the AP-42 Section 1.4. Since this particulate was measured using EPA method 5 (front-half catch), the revised AP-42 will state that the emissions are filterable particulate matter. This filterable particulate emission factor will be given a rating of "B" as dictated by AP-42 Section 1.4.

Primary Reformer (Distillate Oil)

The distillate oil fired primary reformer emits SO_x, NO_x, carbon monoxide (CO), nonmethane organic compounds (NMOC), methane (CH₄), and filterable particulate. Emissions in the 1983 AP-42 are based on this reference and the 1973 AP-42 Section 1.3 Fuel Oil Combustion. Emission factors in AP-42 Section 1.3 are given in terms of mass of emissions per volume of fuel fired for an industrial boiler. This emission factor was then manipulated to reflect emissions in terms of mass of emissions per mass of ammonia produced. Reference 3 provides the combustion rate needed to convert the values given in AP-42 Section 1.3. The combustion rate for distillate oil with heating value of $37x10^6$ Btu/m³ is given for a range of 0.2 to 0.3 cubic meters of fuel per metric ton of ammonia produced. To calculate the revised emission factors using the current AP-42 Section 1.3 (revised October 1986), an average combustion rate of 0.25 cubic meters of fuel per metric ton of ammonia produced was used. This combustion rate is multiplied by the emission factors given in the AP-42 Section 1.3 and any applicable conversion factors to yield the final emission factors which are reported in the revised 1992 AP-42 Section 5.2 Synthetic Ammonia.

The SO_x emitted per kilogram of ammonia produced contains approximately 99 weight percent of SO₂. SO_x emission factor is based on oil containing 0.3 weight percent sulfur. The NO_x emission factor was derived from the testing which indicates that approximately 95 weight percent of the NO_x is NO. NMOC emissions detailed in AP-42 Section 1.3 are lower than the previously reported NMOC emissions. Possible organic emissions from this process include aliphatic and aromatic hydrocarbons, esters, ethers, alcohols, carbonyls, carboxylic acids, and polycylic organic matter. Methane emissions per kilogram of ammonia produced are lowered than the previously reported. The total suspended particulate emissions will be changed to reflect filterable particulate emissions as dictated by AP-42 Section 1.3. The revised 1992 AP-42 will only provide particulate emissions as filterable since no information is available for condensible particulates. All of these revised emission factors will retain their "A" rating as dictated by AP-42 Section 1.3.

Carbon Dioxide Regenerator

The emissions which occur from the carbon dioxide regenerator are carbon monoxide (CO), organic compounds (TOC), ammonia (NH₃), and carbon dioxide (CO₂). Emissions are based on information from seven plants. Material balances and engineering estimates were used at six of these plants. One plant bases its hydrocarbon and CO emissions on unit design calculations. Since

all of these emissions are based on material balances, engineering estimates, or unit design calculations instead of source tests, all of the emission factor ratings will be lowered to an "E." Also, this reference does not include the material balances calculations. The emission factor for CO will remain the same in the revised 1992 AP-42. The NMOC emission factor will be reclassified as total organic compounds (TOC) since the reference does not specify whether the hydrocarbon total given includes methane. The NMOC emissions in the 1983 AP-42 are based on this reference and this reference lists emissions as hydrocarbons and monoethanolamine, not NMOC. In the revised 1992 AP-42, these emissions will be listed as total organic compounds (TOC) and footnoted to contain 0.05 grams of monoethanolamine emitted per kilogram of ammonia produced. The hydrocarbon emissions will be listed as 0.52 grams of TOC emitted per kilogram of ammonia produced. The ammonia emission factor and the CO₂ emission factor will remain the same.

Condensate Stripper

Nonmethane organic compounds (NMOC), ammonia, and carbon dioxide (CO_2) are emitted from the condensate stripper. Emissions are based on 65 source test measurements to determine ammonia and methanol emissions from the steam stripper overheads and seven source test measurements to determine CO_2 emissions. The nonmethane volatile organic compound (NMVOC) emissions given in the 1983 AP-42 will be reclassified as NMOC in the revised 1992 AP-42. The emission factor will be footnoted to indicate that it contains mostly methanol. The ammonia emission factor will remain unchanged, and the carbon dioxide emission factor will be noted to fall within a plus or minus 60 percent range. Since PES does not have these tests for evaluation, the emission factor ratings for all of these emissions will be lowered to "E."

<u>Reference 3</u>: <u>Handbook of Emission Factors.</u>

Emissions in this reference are not given specifically for each step in the synthetic ammonia production process. Instead, a range for overall emissions for the process are presented. No direct comparison to the AP-42 emission factors is possible; however, if the emissions for each step listed in the AP-42 are totalled, then these values can be compared to those presented in this reference. After evaluation, PES concluded that the sum for each type of emission as listed in the 1883 AP-42 fell within the range specified by this reference, except for one. The sum of carbon monoxide emissions from the AP-42 is eight times greater than those given in this reference. This reference does not provide enough information to determine why there is such a large difference between the two factors. Therefore, this reference was not used to change the emissions listed in the 1983 AP-42.

<u>Reference 4</u>: <u>AIRS Facility Subsystem Source Classification Codes and Emission Factor</u> Listing For Criteria Pollutants.

The emission factors presented in AIRS agree with those given in the 1983 AP-42, except for one. AIRS lists the VOC emission factor for an oil fired primary reformer as 0.19 grams of NMOC per kilogram of ammonia produced (0.38 pounds of NMOC per ton). The 1983 AP-42 also lists this emission factor. However, after careful review of the references used to create the AP-42 emission factor, PES concluded that this emission factor should be changed. For details on this revised emission factor, see discussion for Reference 1. The volatile organic compound (VOC) emission factors listed in AIRS are listed under NMOC in the 1983 and 1992 AP-42, since VOC is a subset of NMOC.

4.2 CRITERIA POLLUTANT EMISSIONS DATA

Nonmethane Organic Compounds.

Nonmethane organic compounds (NMOC) can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factor was changed according to the details provided in Chapter 4.1 of this report under the discussion of Reference 2.

Lead.

No information on emissions of this pollutant was found for the synthetic ammonia process.

Sulfur dioxide.

Sulfur oxides can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factor in the 1983 AP-42 was corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2.

Nitrogen oxides.

Nitrogen oxides can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factor in the 1983 AP-42 was corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2.

Carbon monoxide.

Carbon monoxide can be emitted from the synthetic ammonia process. However, since no data on emissions of this pollutant were received, the emission factor in the 1983 AP-42 was corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2.

Total Suspended Particulate & PM₁₀.

Emissions of particulate matter can be divided into three categories: filterable, organic condensible, and inorganic condensible. Filterable particulate matter is that which collects on the filter and in the sampling probe assembly of a particulate sampling train. When emissions testing is performed in accordance with Method 5, the filter and probe are maintained at approximately $120 \,^{\circ}\text{C}$ (248 $^{\circ}\text{F}$); materials that condense at a temperature lower than this will pass through the filter. Many emission tests also quantify emissions of condensible particulate matter, typically that which condenses at or above $20 \,^{\circ}\text{C}$ (68 $^{\circ}\text{F}$). This condensible particulate matter is collected by passing the effluent gas through ice water-cooled impingers such that the gas exiting the last impinger is at a temperature less than $20 \,^{\circ}\text{C}$. The preferred method for quantification of emissions of condensible particulate matter is EPA Reference Method 202. This method entails extraction of the organic portion of the condensible, or back-half, catch with methylene chloride, evaporation of the extract at room temperature, desiccation, and weighing. The inorganic portion of the back-half catch is evaporated at $105 \,^{\circ}\text{C}$ (221 $^{\circ}\text{F}$), desiccated, and weighed.

Particulates can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factor in the 1983 AP-42 was corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2. Only filterable particulate measurements were provided by this reference and no other references were found that contained any further particulate data. No data on PM_{10} emissions are given in any of the references used in the AP-42 or this background report. Therefore, there are no PM_{10} emission factors listed in the AP-42.

4.3 NONCRITERIA POLLUTION EMISSIONS DATA

Hazardous Air Pollutants.

Hazardous air pollutants (HAPs) are defined in the 1990 Clean Air Act Amendments. Methanol and hydrogen sulfide are classified as HAPs and can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factors in the 1983 AP-42 were corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2.

Global Warming Gases.

Pollutants such as methane, carbon dioxide, and nitrous oxide have been found to contribute to overall global warming. For source testing purposes, the concentration of carbon dioxide in the stack gas being tested is measured in order to approximate the molecular weight of the stack gas. This measurement is performed in such a way that its level of accuracy is less than that of the primary pollutants of interest. It is for this reason that the emission factors for CO_2 presented in the AP-42 section are rated no higher than "C."

The preferred method of quantifying emissions of carbon dioxide from combustion processes is through fuel analysis. All of the carbon in the fuel, minus that contained in unburned hydrocarbons and carbon monoxide in the effluent gases, can be assumed to be emitted in the form of carbon dioxide.

Methane and carbon dioxide can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factors in the 1983 AP-42 were corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2.

Ozone Depletion Gases.

Chlorofluorocarbons have been found to contribute to ozone depletion. No information on emissions of these pollutants were found for the synthetic ammonia process.

Other.

Ammonia can be emitted from the synthetic ammonia process. However, since no data on emissions of this pollutant were received, the emission factor reported in the 1983 AP-42 was corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2.

4.4 DATA GAP ANALYSIS

The majority of the emission factors are based on secondary sources such as journals, engineering judgements, and unavailable source tests. Many of the ratings on these factors were lowered for these reasons. The information used to obtain the factors presented in the AP-42 is old and may have changed in the last several years. In order to obtain more reliable emission factors, more source testing needs to be done.

Nonmethane organic compounds (NMOC), methane, carbon dioxide (CO_2), sulfur oxides (SO_x), and nitrogen oxides (NO_x) can be emitted from the synthetic ammonia process. Methanol (MEOH) and hydrogen sulfide (H_2S) are classified as HAPs and can also be emitted from the synthetic ammonia process.No source tests were received which contained data on any of these emissions. The emission factors reported in the revised AP-42 are based on Reference 2. This reference does not present raw source test data; consequently, PES can not verify that these emission factors were calculated correctly in the original source tests. PES can verify that the emissions presented in Reference 2 have been transferred correctly into the revised AP-42.

Particulates can be emitted from the synthetic ammonia process. However, since no data on emissions of these pollutants were received, the emission factor in the 1983 AP-42 was corrected as detailed in Chapter 4.1 of this report under the discussion of Reference 2. Only filterable particulate measurements were provided by this reference and no other references were found that contained any further particulate data. No data on PM_{10} emissions are given in any of the references used in the AP-42 or this background report. Therefore, there are no PM_{10} emission factors listed in the AP-42.

Carbon monoxide (CO) and ammonia (NH₃) can also be emitted from the synthetic ammonia process. Information on these pollutants were received from two facilities. This information could not be classified as complete source tests; therefore, PES could not use the data to update the emission factors. Consequently, the revised AP-42 CO and NH₃ emission factors are based on Reference 2. This reference does not present raw source test data; consequently, PES can not verify that these emission factors were calculated correctly in the original source tests. PES can verify that the emissions presented in Reference 2 have been transferred correctly into the revised AP-42.

In conclusion, PES recommends that further source testing be done for the following pollutants: NMOC, CO, CO₂, NH₃, filterable particulate matter, condensible particulate matter,

 PM_{10} , MEOH, H_2S , CH_4 , SO_x , and NO_x . Based on the summaries above, testing on these pollutants are necessary to develop accurate emission factors for the synthetic ammonia category. Updates to the process description can also be obtained from any new source testing that may be done.

4.5 **REFERENCES FOR CHAPTER 4**

- 1. <u>Source Category Survey: Ammonia Manufacturing Industry</u>, EPA-450/3-80-014, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, August 1980.
- G.D. Rawlings and R.B. Reznik, <u>Source Assessment: Synthetic Ammonia Production</u>, EPA-600/2-77-107m, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, November 1977.
- 3. <u>Handbook of Emission Factors</u>. Ministry of Housing, Physical Planning and Environment, Government Publishing Office, The Hague, Netherlands. 1984.
- 4. <u>AIRS Facility Subsystem Source Classification Codes and Emission Factor Listing For</u> <u>Criteria Pollutants</u>. EPA-450/4-90-003, U. S. Environmental Protection Agency, Research Triangle Park, NC 27711, March 1990.

TABLE 4-4.LIST OF CONVERSION FACTORS

Multiply:	by:	To obtain:
mg/dscm	4.37 x 10 ⁻⁴	gr/dscf
m ²	10.764	ft^2
acm/min	35.31	acfm
m/s	3.281	ft/s
kg/hr	2.205	lb/hr
kPa	1.45 x 10 ⁻¹	psia
kg/Mg	2.0	lb/ton
Mg	1.1023	ton
1	1000	m ³
mg	1000	g
metric ton	0.001	kg

Temperature conversion equations:

Fahrenheit to Celsius:

$$^{\circ}C = \frac{(^{\circ}F - 32)}{1.8}$$

Celsius to Fahrenheit:

 $^{\circ}F = 1.8(^{\circ}C) + 32$

APPENDIX A

AP-42 SECTION 5.2

[Not presented here. See instead current AP-42 Section 8.1]