

SNAP CODE: **010306**

SOURCE ACTIVITY TITLE: **PETROLEUM AND/OR GAS REFINING PLANTS**
Process Furnaces

NOSE CODE: **104.08.03**

NFR CODE: **1 A 1 b**

1 ACTIVITIES INCLUDED

This chapter covers emissions released from combustion processes within a refinery for the heating of crude and petroleum products without contact between flame and products. Primary reduction measures are taken into account (if installed). The emission generating process is the combustion of heavy fuel oil, refinery gas and/or petroleum coke. Thermal cracking units are also taken into account.

The following activities are excluded: power plants installed within a refinery (producing steam and/or electricity) as well as internal combustion engines and gas turbines are considered in chapters B111 on "Combustion Plants as Point Sources" and B112 on "Combustion Plants as Area Sources"; fluid catalytic cracking/CO boilers are treated under SNAP 040102; sulphur recovery plants are covered by SNAP 040103; flaring in the oil industry is treated under SNAP 090204 (waste treatment and disposal). Process specific emissions from refineries are covered by SNAP 040104 "Storage and Handling of Petroleum Products in a Refinery".

2 CONTRIBUTION TO TOTAL EMISSIONS

The contribution of emissions released from refinery process furnaces to the total emissions in countries of the CORINAIR90 inventory is given as follows:

Table 1: Contribution to total emissions of the CORINAIR90 inventory (28 countries)

Source-activity	SNAP-code *	Contribution to total emissions [%]							
		SO ₂	NO _x	NMVOC	CH ₄	CO	CO ₂	N ₂ O	NH ₃
Process Furnaces without Contact	010306	1.4	0.5	0.2	0	0.1	1.0	0.2	-

0 = emissions are reported, but the exact value is below the rounding limit (0.1 per cent)

- = no emissions are reported

* = SNAP90 code 030201

In a modern refinery up to 80 %, in some cases even 90 %, of all SO₂ emissions and also a major part of the NO_x emissions and particulate emissions (combined with heavy metals) of

the refinery are dependent or directly related to the types of fuel used and their respective shares of the total fuel consumption of the refinery /14/. A split of total refinery emissions can be given as an example for SO₂ and NO_x emissions (Western Europe) /cf. 1, 9/:

SO ₂ :	- Process heaters and boilers:	69 %	of total SO ₂ emissions from refineries
	- FCC units (CO boilers):	7 %	”
	- sulphur recovery unit:	10 %	”
	- flares:	9 %	”
	- other sources (e.g. gas turbines, stationary engines):	6 %	”
NO _x :	- process heaters:	46 %	of total NO _x emissions from refineries
	- boilers:	17 %	”
	- FCC units (CO boilers):	16 %	”
	- engines (for pumps etc.):	11 %	”
	- gas turbines and combined cycle systems:	10 %	“

Process heaters contribute about 40 % to the total refinery emissions of SO₂ and NO_x, whereas the contribution of refineries to the total anthropogenic emissions is about 1 % (average for SO₂ and NO_x).

3 GENERAL

3.1 Description

The most relevant emission sources within this sector are process heaters. In most refining processes it is necessary to apply heat to raise the temperature of the feedstock to a required temperature. Process heaters are therefore used, and where processes are self-contained each process usually has its own separate process heater.

3.2 Definitions

Cracking one of the process steps within a refinery for splitting long-chain hydrocarbons into short-chain hydrocarbons. Two types of cracking can be distinguished: catalytic and thermal. Catalytic cracking (e.g. FCC) is the most common type of cracking implemented in refineries. Thermal cracking is of less importance with the exception of visbreaking (thermal cracking of high-boiling residues).

3.3 Techniques

Process heaters are installed as for example pipe still or pre-heaters; they are mostly located at the atmospheric distillation, before the vacuum distillation, before the visbreaker, before the FCC units, before thermal cracking units¹, and before the sulphur recovery units. The burners

¹ Thermal cracking units are of less importance within refineries, but they are commonly used in the petrol chemistry (e.g. olefin cracking units).

are mostly situated at the bottom of the installations. Refineries can have about 4 up to more than 40 process heaters depending on the complexity of operations. The refinery gas, produced by petroleum processing, provides a significant part of the fuel for process heaters. Different processes contribute varying amounts and varying compositions to the refinery gas. However, major components of the gas are hydrogen and light hydrocarbons. In principle, refineries use gaseous fuels (refinery gas, sometimes also natural gas /14/), which are supplemented by liquid fuels (heavy fuel oil or other residues) and solid fuels (petroleum coke). In many applications, dual-fuel burners are used with gas and liquid fuel being consumed by the same burners. The columns can also be heated by using the process steam generated in boilers. The fuel used for steam generation may be different from commercial fuel as its nature is determined by the optimal use of resources within each refinery at a given time. /cf. 1, 3/.

3.4 Emissions

Relevant pollutants are sulphur oxides (SO_x), nitrogen oxides (NO_x) and carbon dioxide (CO_2). SO_3 emissions are negligible for all fuels used. For normal operating conditions emissions of carbon monoxide (CO), nitrous oxide (N_2O), and heavy metals are of less relevance. Emissions of volatile organic compounds (non-methane VOC and methane (CH_4)) can occur, but they are often negligible. Normally, emissions of ammonia (NH_3) are not relevant.

Emissions considered here are associated with continuous operation of the refinery. Emissions are released through stacks. Nevertheless, frequent start-ups and shut-downs of process heaters may occur, due to unexpected changes in operating conditions or from regular non-operating times (e.g. for maintenance).

The emissions of sulphur dioxide (SO_2) are directly related to the sulphur content of the fuel(s) used. The sulphur content of refinery gas varies from 0.01 to 5 %, averaging 0.8 % /2/. Refinery gases are produced in almost all hydrocarbon processing installations. They can be classified as sulphur-free gases and gases containing sulphur: /14/

- sources of sulphur-free gases: ²
 - isomerisation plants, catalytic reforming plants, hydrogen manufacturing plants, gasification of coke in some coke operations,
- sources of sulphur-containing gases: ³
 - crude distillation, hydro-treating/hydro-desulphurisation, catalytic cracking, thermal cracking/coking/visbreaking, residue conversion, flare gas recovery⁴, gasification of coke from some coke operations /14/ ⁵

² As a consequence of the use of sulphur sensitive catalysts in isomerisation and catalytic reforming, these processes require virtually sulphur free feedstocks. As a result, the gas streams from these units are nearly sulphur free. The gases produced in hydrogen manufacturing plants and from gasification units can also be desulphurised. /cf. 14/

³ Most other gases produced in the refinery contain hydrogen sulphide (H_2S) and often small quantities of mercaptans.

Liquid fuels used in a refinery originate from various processes (e.g. crude oil distillation, high vacuum distillation, thermal or catalytical cracking). In general, the liquid fuels comprise the following components: atmospheric and vacuum residues, thermally cracked residues, heavy catalytically cracked cycle oil and hydrocracked residues. Sulphur contents of liquid refinery fuels and/or components are given in Table 2. /14/

Table 2: Sulphur contents of liquid refinery fuels (cf. /14/)

Residue	Sulphur content of residues [wt.-%] from	
	Crude oil from North Sea	Crude oil from Middle East
Atmospheric residue	0.6 - 1.1	2.3 - 4.4
Vacuum residue	1.1 - 1.8	3.6 - 6.1
Cracked residue	n. d.	3.5 - 6.5

n. d. : no data are available

However, lower amounts of sulphur in liquid refinery fuels may occur (e.g. Swedish refineries use fuel oil with a sulphur content of 0.4 - 0.5 wt.-% /cf. 15/). An average sulphur content of fuel oil used in refineries is given as 2.8 wt.-% in /cf. 2/. A weighted average sulphur content of the mix of refinery gas and heavy fuel oil can be given as 1.7 wt.-% /2/.

The sulphur content of the petroleum coke produced and consumed by refineries depends on the type of crude oil/fractions used. In practice, the sulphur content of coke varies between ca. 0.93 wt.-% sulphur (petroleum coke produced mostly from delayed coking process) and ca. 1.4 wt.-% sulphur (petroleum coke produced mostly from fluid coking process) /cf. 6/.

The formation of nitrogen oxides (NO_x) can be split into “fuel-NO”, “thermal-NO” and “prompt-NO” as discussed in chapter B111 on “Combustion Plants as Point Sources” (Section 3). Within the temperature range under consideration, the formation of “prompt-NO” can be neglected. “Fuel-NO” results from the oxidation of the fuel nitrogen content. For liquid fuels, the content of nitrogen in heavy fuel oil varies between 0.1 and 0.8 wt.-% /7/. The content of nitrogen in gaseous fuels (refinery gas) as well as in solid fuels (petroleum coke contain ca. 0.2 wt.-% /6/) is negligible. Relatively high NO_x emissions may be released by thermal cracking units in petrochemical industry (e.g. at olefin cracking units concentrations of about 130 up to 1,600 mg/m³ may occur /18/). The design of burner and furnace as well as the operating conditions determine the NO_x formation. NO_x emissions vary considerably for existing and new (optimised) furnaces in refineries (see Table 3).

⁴ The flare gas recovery system is in fact a refinery safety device which under normal conditions has no flow /14/.

⁵ For a description of selected units see chapter B411 on "Petroleum Products Processing".

Table 3: Ranges of NO_x concentrations for existing and new (optimised) furnaces in refineries (according to /14/)¹⁾

Type of process furnace	NO _x concentration [mg/Nm ³]	
	Gas ²⁾	Refinery fuel ³⁾
Existing furnace	160 - 1,300	280 - 1,000
Furnace with optimal burner and furnace design	100 - 200	about 250 ⁴⁾

1) Apart from firing either gas or liquid fuel separately, mixed gas/liquid firing in the same furnace is also practised in refineries, and emission values may differ considerably from the values observed in the case of gas fired units /14/.

2) The lower range relates to natural gas firing /14/.

3) Thermally cracked residue /14/; liquid fuel

4) However, low-NO_x-burners are reported not to be available for low grade liquid fuels. /cf. 14/

Emissions of carbon monoxide (CO), methane (CH₄) and non-methane volatile organic compounds (NMVOC) are mainly associated with poorly regulated combustion processes; they are small when processes are managed correctly.

Heavy metal emissions are mainly determined by the type of fuel used; only liquid and solid fuels are of relevance. Most of the heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, Se, Zn, V) are normally released as compounds (e.g. chlorides) in association with particulates (see also chapter B111 on “Combustion Plants as Point Sources”, Section 9). In the case of heavy fuel oil mainly Ni and V are of relevance. Particulate emissions originate from two different sources within a refinery; firstly from process heaters and boilers and secondly from FCC units (CO-boilers), which are not considered here. Their contribution to the total emissions is roughly equal. /cf. 8/.

3.5 Controls

SO₂ emissions from process furnaces are only controlled by the use of low sulphur fuels (e.g. by switching from liquid fuels to gaseous fuels, which contain less sulphur).

For the control of NO_x emissions from process furnaces only primary measures are installed (e.g. low-NO_x-burner, flue gas recirculation). The reduction efficiencies for low-NO_x-burners vary between 10 and 30 %, and for flue gas recirculation between 5 and 15 %. At thermal cracking units in petrochemical industry (e.g. olefine cracking units) also secondary abatement measures may be installed (e.g. SCR, SNCR) /18/.

4/5 SIMPLER AND DETAILED METHODOLOGY

Here both approaches refer to the calculation of emissions based on emission factors and activities, which are jointly discussed in the following. The “simpler methodology” is considered as an overall approach, where activity data refer to production figures. The “detailed methodology” is considered as the recommended approach, where activity data concerning the fuel consumption in refinery process furnaces is available for individual plants. The simpler and the detailed methodologies cover all relevant pollutants.

The annual emission is determined according to Equation (1) by an activity and an emission factor:

$$E_i = EF_i \cdot A \quad (1)$$

E_i annual emission of pollutant i
 EF_i emission factor of pollutant i
 A activity

The activity A and the emission factor EF_i have to be determined on the same level of aggregation by using available data. The CORINAIR90 methodology requires for refinery process furnaces activity data, which is related to the type of fuel consumed in [GJ/a].

4.1 Simpler methodology

The simpler methodology corresponds to an approach, which takes into account activity rates derived from data of comparable installations or from literature data. Here, it is assumed, that the required activity data (according to CORINAIR90) are not available (see Equation (1)). In practice, statistical material (see also Section 6), which often provides only the throughput of crude oil in [Mg/a], has to be used. Some national statistics publications also provide throughputs per individual refinery.

In order to approximate activity data referring to the energy input into process heaters in [GJ/a] the specific energy consumption has to be taken into account as given e.g. in Equation (2):

$$A_{COR} = F \cdot A_{Stat} \quad (2)$$

A_{COR} activity in CORINAIR-compatible unit (energy input [GJ])
 F specific energy consumption (energy input/mass crude oil [GJ/Mg])
 A_{Stat} activity directly obtained from statistics (mass crude oil [Mg])

For the determination of the specific energy consumption F , related to the throughput of crude oil, only the own consumption of the refinery has to be taken into account. The own consumption of a refinery amounts to about 5 % (average in 1990) of the input (crude oil and intermediate products) /3, 5/. For hydroskimming refineries⁶ the fuel demand may vary between 2 and 3 wt.-% and for complex, high conversion refineries between 6 and 8 wt.-% /14/. About 40 % of the refinery fuel consumption is used for process heaters. The fuel split of refinery fuel for its own consumption can be given as: heavy fuel oil (ca. 35 %), petroleum coke (ca. 10 %) and refinery gas (ca. 55 %); the relevance of gas oil is < 1% and can be neglected /3/.

However, the simpler approach leads to significant uncertainties. Therefore, no emission factors are provided for this edition.

⁶ The simplest type of refineries, the so-called „hydro-skimming“ refineries, carry out very little conversion into various products. The product distribution is largely determined by the composition of the crude oil processed and cannot be influenced to a great extent by modifying the operating mode of the refineries. /14/

4.2 Detailed methodology

The detailed methodology corresponds to a plant specific approach, which takes into account as far as possible plant specific information. Here, CORINAIR90 compatible activity data for refinery process furnaces (related to the type of fuel consumed in [GJ/a]) are directly available (Equation (1)).

The following two sections provide individual approaches for the determination of SO₂ and CO₂ emission factors.

4.2.1 SO₂ emission factors

Emission factors for SO₂ in [g/GJ] are given in Table 6 (see Section 8) based on literature data. SO₂ emissions can be directly correlated to the sulphur content of the fuel and the fuel consumption. Emission factors for SO₂ in [g/GJ] can be obtained by using Equation (2):

$$EF_{SO_2} = 2 \cdot C_{S_{fuel}} \cdot \frac{1}{H_u} \cdot 10^6 \quad (2)$$

EF_{SO₂} emission factor of SO₂ [g/GJ]

C_{S_{fuel}} sulphur content of fuel [wt.-%]

H_u lower heating value [MJ/kg]

If no data is available, default values are recommended:

- sulphur content of fuel: see Section 3.4,
- lower heating value: see Table 4.

Table 4: Lower heating values of refinery fuels

Fuel used	NAPFUE code	Lower heating value [MJ/kg] /10/
Petroleum coke	110	29.31
Gas oil	204	42.70
Heavy fuel oil	203	41.03
Refinery gas	308	48.36

4.2.2 CO₂ emission factors

Emission factors for CO₂ are given in Table 6 (Section 8) based on literature data. Own estimations can be made according to Equation (3) and by using an approximation for the composition of oil, gas and coke as given in Table 5:

$$EF_{CO_2} = \frac{M_{CO_2}}{M_i} \cdot \frac{1}{H_u} \cdot 10^6 \quad (3)$$

EF_{CO₂} emission factor for CO₂ [g/GJ]

M_{CO₂} molecular weight of CO₂ [g/mol]

M_i molecular weight of fuel i (see Table 5) [g/mol]

H_u lower heating value [MJ/kg]

Table 5: Approximations for the molecular weight /3/

Fuel	Approximation	Molecular weight [g/mol]
Fuel oil	HC	13
Petroleum coke	C	12
Refinery gas	CH ₄ /C ₂ H ₆	23 ¹⁾

¹⁾ An assumption concerning the average molecular weight has been made as both fractions are included with a mass portion of 50:50.

4.2.3 Emission factors for other pollutants

Emission factors for the pollutants NO_x, CH₄, NMVOC, CO, and N₂O are given in Table 6 (see Section 8) based on literature data depending on the type of fuel used. Emission factors for heavy metals are given in Table 7 (see Section 8).

6 RELEVANT ACTIVITY STATISTICS

The following statistics can be used for the determination of the throughput of crude oil. The consumption of intermediate products has to be taken into account separately. In some national statistics also the total own consumption of fuels within the refineries is reported.

- Statistical Office of the European Communities (EUROSTAT) (ed.): CRONOS Databank; 1993 (Crude oil production XX 93 0603 3, Crude oil treated in refineries XX 93 0604 3)
- Office for Official Publication of the European Communities (ed.): Annual Statistics 1990; Luxembourg; 1992

Information concerning European refineries is also provided by Concawe (Den Haag).

7 POINT SOURCE CRITERIA

Refineries have to be treated as point sources according to the CORINAIR90 methodology. Process furnaces within a refinery have to be reported collectively as a part of a refinery.

8 EMISSION FACTORS, QUALITY CODES AND REFERENCES

The following Table 6 contains emission factors for selected pollutants based on literature data.

Table 6: Emission factors of gaseous pollutants for process heaters in refineries¹⁰⁾

Type of fuel			NAPFUE code	Emission factors						
				SO ₂ [g/GJ]	NO _x [g/GJ]	NMVOC [g/GJ]	CH ₄ [g/GJ]	CO [g/GJ]	CO ₂ [kg/GJ]	N ₂ O [g/GJ]
s ¹⁾	coke	petroleum	110		300 ⁷⁾				101 ⁵⁾	22 ⁷⁾ g/Mg
l ²⁾	oil	residual	203	245 - 1,962 ⁹⁾	100 - 210 ⁸⁾ 75 - 328 ⁹⁾	1 - 41 ⁹⁾	0.1 - 3.5 ⁹⁾	7 - 350 ⁹⁾	78 ⁵⁾ 53 - 79 ⁹⁾	22 ⁷⁾ g/Mg 2 - 22 ⁹⁾
G	gas	natural	301	0.7 - 432 ⁹⁾	1.4 - 140 ⁹⁾	0.3 - 7 ⁹⁾	0.3 - 4 ⁹⁾	1.3 - 280 ⁹⁾	53 - 55 ⁹⁾	1.5 - 22 ⁹⁾ ,
G ³⁾	gas	liquified petroleum	303	1.7 ⁹⁾		14 ⁹⁾	6 ⁹⁾	45 ⁹⁾	64 ⁹⁾	1.5 ⁹⁾
G	gas	refinery	308	12.5 - 1,423 ⁹⁾	90 ⁸⁾ , 140 ⁷⁾ , 155 ⁶⁾ , 30 - 150 ⁸⁾ 35 - 756 ⁹⁾	0.3 - 10 ⁹⁾	0.3 ⁶⁾ 0.3 - 4 ⁹⁾	280 ⁶⁾ 10 - 280 ⁹⁾	60 ^{4), 6)} 10 - 57 ⁹⁾	1.5 ⁶⁾ , 0.3 - 22 ⁹⁾ , 22 ⁷⁾ g/Mg
l/g	mixtur	of oil/gas	-	220 ⁴⁾ g/Mg	350 ⁴⁾ g/Mg					

- 1) In CORINAIR90 also NAPFUE codes 103 and 105 have been reported
2) In CORINAIR90 also NAPFUE code 204 has been reported
3) In CORINAIR90 also NAPFUE code 303 has been reported
4) CONCAWE /9/, range for SO₂ emission factors: 70 - 430 g/Mg
5) BMU, Germany 1993 /12/
6) BMU, Germany 1994 /16/
7) CORINAIR /4/
8) CONCAWE /1/
9) CORINAIR90 data
10) At this stage emission factors for thermal cracking units are not available.

Table 7: Heavy metal emission factors for gaseous fuels fired in refinery process heaters /13/

Source	Emission factor [g/TJ]			
	Cr (total)	Cr ⁶⁺ (Hex) ¹⁾	As	Cd
Process heater:				
- Single stage	0.10	0.19	0.03	0.01
- With LNB	0.05	0.01	0	0
- With air preheater	0.05	0	0	0.14
- Reformer	0.09	0.19	0	0

¹⁾ Cr⁶⁺ is reported separately due to the high toxicity of this species.

For refinery process heaters, which are fed with fuel oil, emission factors for heavy metals are approximately the same as given in Table 31 in chapter B111 on "Combustion Plants as Point Sources". Residues fuelled in refineries need not be comparable to heavy fuel oils as a product. Therefore, composition data of residues are needed in order to estimate heavy metal emissions /17/.

9 SPECIES PROFILES

Species profiles (oxides of sulphur and nitrogen) are comparable to those released from combustion installations. Details can be found in chapter B111 "Combustion Plants as Point Sources" (Section 9).

10 UNCERTAINTY ESTIMATES

At this stage no information is available.

11 WEAKEST ASPECTS/PRIORITY AREAS FOR IMPROVEMENT IN CURRENT METHODOLOGY

Weakest aspects discussed here are related to emission factors and activities.

Data provided by CORINAIR90 project can only be used in order to give a range of emission factors. Further work should be invested to develop emission factors which take into account technical specifications and fuel characteristics.

Most refineries process crude oil as well as intermediate products coming from other refineries. The energy demand for the processing of intermediate products differs from the energy demand for the processing of crude oil. The share of intermediate products processing varies strongly. Therefore, further work should be invested in providing characteristic profiles for the energy consumption (own consumption) of a refinery, which are suitable for inventorying purposes.

12 SPATIAL DISAGGREGATION CRITERIA FOR AREA SOURCES

13 TEMPORAL DISAGGREGATION CRITERIA

Temporal disaggregation of annual emission data (top-down approach) provides a split into monthly, weekly, daily and/or hourly emission data. Temporal disaggregation of annual emissions released from process furnaces in refineries can be obtained by taking into account the:

- time of operation and
- variation of load.

Data for the annual time of operation in refineries is available from statistics. In principle, refineries produce continuously during the whole year except during standstill time due to maintenance.

The load of the refinery is determined by the variation of production due to the varying demand for petroleum products. Information concerning the variation in the production or the

demand for refinery products can only be obtained directly from refinery operators. (Note: the short time demand for refinery products is met by refinery products being stored in tanks.).

14 ADDITIONAL COMMENTS

15 SUPPLEMENTARY DOCUMENTS

16 VERIFICATION PROCEDURES

As outlined in the chapter on "Concepts for Emission Inventory Verification" different verification procedures can be recommended. Verification procedures considered here are principally based on the verification of emission data on a national level and on a plant level. The verification on a plant level relies on comparisons between calculated emissions/emission factors and those derived from emission measurements.

17 REFERENCES

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18 BIBLIOGRAPHY

19 RELEASE VERSION, DATE AND SOURCE

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20 POINT OF ENQUIRY

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Abbreviations

FCC	Fluid Catalytic Cracking
g	gaseous state of aggregation
l	liquid state of aggregation
LNB	Low-NO _x -Burner
s	solid state of aggregation
SCR	Selective Catalytic Reduction
SNCR	Selective Non-Catalytic Reduction