

Eemeli Tsupari, Suvi Monni & Riitta Pipatti

# Non-CO<sub>2</sub> greenhouse gas emissions from boilers and industrial processes

Evaluation and update of emission factors for the Finnish national greenhouse gas inventory



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ISBN 951–38–6759–5 (soft back ed.) ISSN 1235–0605 (soft back ed.) ISBN 951–38–6760–9 (URL: http://www.vtt.fi/inf/pdf/) ISSN 1455–0865 (URL: http://www.vtt.fi/inf/pdf/) Copyright © VTT 2005

JULKAISIJA – UTGIVARE – PUBLISHER

VTT, Vuorimiehentie 3, PL 1000, 02044 VTT puh. vaihde 020 722 111, faksi 020 722 4374

VTT, Bergsmansvägen 3, PB 1000, 02044 VTT tel. växel 020 722 111, fax 020 722 4374

VTT Technical Research Centre of Finland, Vuorimiehentie 3, P.O.Box 1000, FI–02044 VTT, Finland phone internat. +358 20 722 111, fax +358 20 722 4374

VTT Prosessit, Biologinkuja 7, PL 1602, 02044 VTT puh. vaihde 020 722 111, faksi 020 722 7026

VTT Prosesser, Biologgränden 7, PB 1602, 02044 VTT tel. växel 020 722 111, fax 020 722 7026

VTT Processes, Biologinkuja 7, P.O.Box 1602, FI–02044 VTT, Finland phone internat. +358 20 722 111, fax +358 20 722 7026

Technical editing Anni Kääriäinen

Tsupari, Eemeli, Monni, Suvi & Pipatti, Riitta. Non-CO<sub>2</sub> greenhouse gas emissions from boilers and industrial processes. Evaluation and update of emission factors for the Finnish national greenhouse gas inventory. Espoo 2005. VTT Tiedotteita – Research Notes 2321. 82 p. + app. 24 p.

**Keywords** greenhouse gases, emission factors, energy production, stationary combustion, co-firing, industrial processes, nitrous oxide, volatile organic compounds, methane, carbon monoxide, recommendations, CO, CH<sub>4</sub>, N<sub>2</sub>O, NMVOC

## Abstract

Finland is obligated to report anthropogenic greenhouse gas emissions annually to the UNFCCC. Inventories of greenhouse gas emissions and methodologies used have to be well based and documented. The aim of this report is to give input to Finnish greenhouse gas inventory of energy (stationary combustion) and industrial sectors. The emission factors for the industrial processes used in the inventory are based both on plant-specific factors and the IPCC default values. The currently used emission factors for the non-CO<sub>2</sub> greenhouse gases from combustion are mainly based on studies done at the beginning of 1990's, even though small modifications to the emission calculation model have been done afterwards. Since 1990, significant changes have occurred in plant design and maintenance, operating conditions and composition of fuels. In addition, knowledge of emission generating processes has increased world-wide. In this study, the non-CO<sub>2</sub> emission factors (mainly CH<sub>4</sub> and N<sub>2</sub>O, and at a lesser extent, CO and NMVOC) for combustion and industrial processes used in the Finnish greenhouse gas inventory are evaluated based on domestic and international literature, available measurement data and expert interviews. Results of this study revealed many potential areas for improvements in the inventory. Consequently, recommendations to update the CH<sub>4</sub> and N<sub>2</sub>O emission factors from boilers and industrial processes are given including guidance on the level of the calculations. The time dependency and uncertainties of the emission factors are also considered. In addition, effect of load and fuel mixes on emissions is examined. Areas for further research are also indicated.

# Preface

The aim of this report was to evaluate the non- $CO_2$  emission factors used in the Finnish greenhouse gas inventory under the UNFCCC. The sectors covered were Energy (excluding transportation) and Industrial Processes, and gases  $CH_4$  and  $N_2O$  (also CO and NMVOC at a lesser extent). The project was funded by Statistics Finland. Co-operation with Mirja Kosonen and Kari Grönfors from the funding organisation enabled successful completion of the work, of which the authors want to express their warmest thanks.

Matti Nuutila from Finnish District Heating Association and Liisa Aarikka and Olli Kallio from Porin Lämpövoima Oy provided important information about partial load. Arto Riikonen from Gasum gave useful information on emissions from natural gas combustion. Jukka Leskelä from Finergy and Pia Kilpinen from Åbo Academi are also acknowledged for giving input and comments to the work.

The authors wish also to thank various researchers at VTT, who lent their expertise for this work. Jukka Lehtomäki, Kauko Tormonen and Tuula Vahlman provided information on emission measurement techniques and evaluated recommended emission factors in the report. Raili Vesterinen, Martti Aho, Veli Linna and Heikki Oravainen gave important information especially on emission measurements of peat and biomass combustion. Lars Kjäldman and Marja Keinonen provided information on the processes and factors influencing the emissions. Matti Kytö, Seppo Niemi, Raimo Turunen and Kari Mäkelä gave important information especially on diesel and dual-fuel engines.

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Appendix D: CO from stationary combustion

Appendix E: NMVOC from stationary combustion

# List of abbreviations

ABFB	atmospheric pressure bubbling fluidized bed
ACFB	atmospheric pressure circulating fluidized bed
BFB	bubbling fluidized bed
BFBC	bubbling fluidized bed combustion/combustor
CaO	calcium oxide
CFB	circulating fluidized bed
CFBC	circulating fluidized bed combustion/combustor
$CH_4$	methane
СНР	combined heat and power
CO	carbon monoxide
$CO_2$	carbon dioxide
CRF	common reporting format (used in the reporting to the UNFCCC)
$C_xH_y$	hydrocarbon
d	diameter
DBB	dry bottom boiler
EF	emission factor
FBC	fluidised bed combustion/combustor
FGD	flue gas desulphurization
FID	flame ionisation detector
FTIR	Fourier transform infrared spectroscopy
GC	gas chromatography
GFC-IR	gas filter correlation infrared technique
GHG	greenhouse gas
GTCC	gas turbine combined cycle
H <sub>2</sub> O	water
HCN	hydrocyanic acid
HFO	heavy fuel oil
HHV	higher heating value
HPLC	high performance liquid chromatography
IC	ion chromatography
IEF	implied emission factor (typically more aggregated data than EF)
IGCC	integrated gasification combined-cycle

IPCC	Intergovernmental Panel on Climate Change
LFO	light fuel oil
LHV	lower heating value
LPG	liquefied petroleum gas
m	mass
MSW	municipal solid waste
Ν	nitrogen
$N_2$	molecular nitrogen
NDIR	non-dispersive infrared technique
NH <sub>3</sub>	ammonia
$N_2O$	nitrous oxide (dinitrogen oxide)
NMVOC	non-methane volatile organic compounds
NO	nitrogen monoxide
NO <sub>x</sub>	nitrogen oxide
NSCR	non-selective catalytic reduction (NOx reduction technique)
O <sub>2</sub>	molecular oxygen
OFA	over fire air systems
PC	pulverised combustion
PF	pulverized firing (same as PC)
ppm	parts per million
PFBC	pressurized fluidized bed combustion
RDF	refuse derived fuel
REF	recycled fuel
SCR	selective catalytic reduction (NO <sub>x</sub> reduction technique)
SNCR	selective non-catalytic reduction (NOx reduction technique)
$SO_2$	sulphur dioxide
t	ton
tds/d	tons of dry solids per day
TOC	total organic carbon
UNFCCC	United Nations Framework Convention on Climate Change
VAHTI	Regional Environment Centres' database including annual emission register
VOC	volatile organic compounds
WBB	wet bottom boiler

## 1. Introduction

Finland has prepared greenhouse gas inventories under the United Nations Framework Convention on Climate Change for the years 1990 to 2002 (Ministry of the Environment, 2004). The emission factors for the non-CO<sub>2</sub> greenhouse gases from combustion used in the inventory are mainly based on studies done at the beginning of 1990's (Boström et al., 1990; Boström et al., 1992; Boström, 1994). These studies complemented the Finnish energy statistics with an emission calculation system. The studies included the development of a boiler database with information on the combustion technology, fuel and/or fuel mix and emission factors. The emission factors for the non-CO<sub>2</sub> greenhouse gases were based on evaluation of the scientific literature and research on factors influencing the emissions. Results from plant scale measurements and laboratory scale experiments were used in the evaluation (e.g. Dahlberg et al., 1988; Andersson et al., 1989; Cooper et al., 1991; Karlsson & Gustavsson, 1991). The emission factors represented best estimates for country-specific factors for Finland at that point of time.

Since the beginning of 1990, significant changes have occurred in plant design and maintenance, operating conditions and composition of fuels. For example combustion control by measurements from flue gases and different NO<sub>x</sub>-control techniques have become more general in power plants. In addition, knowledge of emission generating processes has increased world-wide. Consequently, the calculation system for the energy based emissions in Finland's national greenhouse gas inventory has been developed further, but the emission factors for the non-CO<sub>2</sub> greenhouse gases have remained largely constant throughout the period, with some exceptions. New plants types not covered by the studies by Boström (1994) have been included in the boiler database and the calculation system. The emission factors for these technologies have been determined through interpolation, or extrapolation when the plant size has been new, or using expert judgement when the fuel mix and/or technologies have changed. A measurement project by Korhonen et al. (2001) gave also new information on emission factors for some technologies. The updates of the database have not always been systematic.

The emission factors for the industrial processes are based both on plant-specific factors and the IPCC default values. Plant-specific values based on measurements are available for nitric acid production in Finland. CH<sub>4</sub> emissions have also been measured at iron and steel production plants.

In this study, the non- $CO_2$  emission factors for combustion and industrial processes used in the Finnish greenhouse gas inventory are evaluated. The study covers only emissions from combustion and processes, i.e. other related emissions from, e.g. fuel production, transmission and distribution are not included. Emission factors are evaluated based on currently available information on the emissions using domestic and international literature, available measurement data and expert interviews as sources. Recommendations to improve and update the emission factors are given including guidance on the level of the calculations. The time series of emission factors is also considered, because it is supposed that due to, e.g. changes in plant design and maintenance emission factors may be different for the base year of the Kyoto Protocol (1990) and the current year.

The non- $CO_2$  emissions depend on plant-specific factors like the technology, fuel properties and process conditions. It is therefore difficult to give generally applicable emission factors. Representative measurement data for specific plant types and processes would give the best basis for emission estimates but are rarely available because usually measurement periods are short and therefore the results do not represent annual average. The implementation of extensive measurement programmes to improve the emission factors cannot however be justified due the small significance of these emissions in the national greenhouse gas inventory, the relatively high cost of the measurement needs have therefore been estimated in context to importance of the emission sources to the inventory.

CH<sub>4</sub> and N<sub>2</sub>O emissions from stationary combustion are discussed and presented in Chapter 2 by gas, fuel and technology. Point sources included in the Finnish boiler data base (CRF<sup>1</sup> categories 1A1 and 1A2) and small-scale combustion (CRF category 1A4) are both presented. Recommendations for emission factors to be used in the Finnish GHG inventory are given in Section 2.3. The analysis of the significance of the different emissions sources and recommendations for further research are given in Chapter 3. Emission factors for carbon monoxide (CO) and non-methane volatile organic compounds (NMVOC) from stationary combustion are also presented, but in a more superficial way in Appendices D and E. CH<sub>4</sub> and N<sub>2</sub>O emissions factors for Industrial processes (CRF category 2) are addressed in Chapter 4 accompanied with a list of possible new emission sources. The discussion and conclusions are in Chapter 5.

<sup>&</sup>lt;sup>1</sup> CRF refers to Common Reporting Format. Annex I Parties to the UNFCCC are reporting their national greenhouse gas emissions and removals to the Convention using so-called CRF Tables. The tables are based on a classification system for reporting GHG emissions by Sector, sink/source categories and gas developed by the IPCC. The common reporting framework includes definitions for the categories and fuels (IPCC, 1997).

## 2. Stationary combustion – CH<sub>4</sub> and N<sub>2</sub>O

#### 2.1 Introduction

Stationary combustion covers over 60% of Finland's greenhouse gas (GHG) emissions, carbon dioxide (CO<sub>2</sub>) being the most important contributor. Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) emissions are smaller in magnitude than the CO<sub>2</sub> emissions. The CO<sub>2</sub> emissions from stationary combustion are known relatively accurately, whereas the uncertainties in the CH<sub>4</sub> and N<sub>2</sub>O emission estimates are large.

CH<sub>4</sub> emissions from stationary combustion contributed less than 1% and N<sub>2</sub>O about 2% to the total CO<sub>2</sub>-equivalent GHG emissions from stationary combustion in Finland in 2002 (Ministry of the Environment, 2004). CH<sub>4</sub> emissions from biomass combustion (reported in Other Sectors [1A4] of the CRF) and N<sub>2</sub>O emissions from peat combustion (reported under other fuels in Energy Industries [1A1]) are the most important sources of these emissions under stationary combustion in Finland. These were also identified as key sources<sup>2</sup> in the Finnish Greenhouse Gas Inventory for the year 2002.<sup>3</sup> In addition, N<sub>2</sub>O from biomass combustion in Energy Industries was identified as a key source by trend assessment. These emissions are small (0.27 Gg N<sub>2</sub>O in 2002), but have grown substantially during the time period from 1990 to 2002 (tenfold in 2002).

#### 2.1.1 Formation of $CH_4$ and $N_2O$ in combustion

Fuel type, combustion technology and conditions during the combustion have a significant effect on the  $CH_4$  and  $N_2O$  emissions. Important factors are temperature, air ratio and residence time in the furnace.

Main reasons for methane emissions from stationary combustion are poor mixing and early quenched oxidation reactions in the combustion process. The same factors have also an effect on CO emissions, and therefore correlation between  $CH_4$  and CO emissions can often be found. Conditions where methane is emitted to the atmosphere are rare in large furnaces, but common in the small combustion devices, such as heating

 $<sup>^{2}</sup>$  A key source category is one that is prioritised within a national inventory system because its estimate has a significant influence on the country's total inventory of direct greenhouse gases in term of the absolute level of emissions, trend in emissions, or both. Large uncertainty in emission source may also raise it as a key source (IPCC, 2000).

<sup>&</sup>lt;sup>3</sup> Key sources and shares of different fuels presented in this Chapter are based on emission inventory of 2002. If emission factors recommended in this report (Chapter 2.3) are taken at use, these may change.

stoves, open fireplaces etc. (Boström et al., 1991). In Finland, nearly 80% of the CH<sub>4</sub> emissions from stationary combustion derive from small-scale combustion (CRF category 1A4) whereas the fuel combusted in small-scale applications is less than 15% of all fuel use in stationary combustion (CRF categories 1A1, 1A2, 1A4 and 1A5).

The combustion process can be divided into four stages: (i) drying of the fuel (water is released as steam), (ii) pyrolysis or release of volatile compounds from the fuel, (iii) ignition and burning of the volatile compounds (formation of coke and combustion gases) and (iv) burning of the coke to ash.  $CH_4$  is released from the fuel in the devolatilization phase (ii), and oxidised to  $CO_2$  in the furnace. Effective oxidation requires high temperatures and sufficiently high air ratio. In low temperatures (< 800 °C) and reducing environments the oxidation reactions are slower (Kilpinen, 2002a), and higher methane emissions might occur.

The fuel type is also of importance to the emissions. For example for brown coal, CH<sub>4</sub> emission factors may be several times higher than for hard coal (IPCC, 1997). Partial load may increase CH<sub>4</sub> emissions due to decreased temperature and loss of optimal combustion conditions, because optimal conditions are usually designed for full load. Some emission control techniques like after-burners may reduce methane emissions (but natural gas firing burners may also increase emissions if poor mixing occurs).

The amount of excess air might have a great effect on the  $CH_4$  emissions. Some  $NO_x$ -reducing technologies like air staging, over fire air systems (OFA) and low- $NO_x$  burners could also be taken into account when estimating the emissions. According to IPCC (1997), none of the emission control technologies (SCR, OFA, low- $NO_x$  burners, ammonia injection, etc.) has a significant effect on methane emissions in utility and industrial scale, but some measurements show that significant reduction of  $CH_4$  emissions can occur in SCR. Because methane emissions vary largely with plant size, combustion technology and conditions (e.g. air ratio and fuel characteristics), accurate estimation of average annual emission factors for plants is difficult and would require very detailed information on the above mentioned factors. There is plenty of information available on  $CH_4$  emission factors in literature. Some of these factors diverge significantly from others due to e.g. different basis of the factors (e.g. heating value used and conditions in the measured plant).

Formation of  $N_2O$  depends on the fuel type, nitrogen content of the fuel and the operating conditions, especially on temperature, fraction of excess air and catalytic activity of char and mineral matter (Korhonen et al., 2001; Löffler et al., 2002; Kilpinen, 2002b). CO-content in furnace may also have an effect on  $N_2O$  formation (Åmand et al., 1991).

There are different opinions about the importance of the different N<sub>2</sub>O formation paths. N<sub>2</sub>O is formed from volatile cyano and cyanide compounds (e.g. HCN) originating from fuel nitrogen but also from nitrogen of char and by heterogeneous reactions on char surface. Formation from HCN is often presented as main formation path but for example NO conversion to N<sub>2</sub>O on char surface may play a significant role (Åmand et al., 1991). Ammonia (NH<sub>3</sub>) from fuel forms mainly NO<sub>x</sub> and N<sub>2</sub>. Molecular nitrogen (N<sub>2</sub>) and oxygen (O<sub>2</sub>) need higher temperatures to react among themselves and therefore they form mainly NO<sub>x</sub> (in temperatures > 1200 °C). Therefore, nitrogen of air and natural gas do not have an effect on N<sub>2</sub>O emissions due to molecular form.

Nitrous oxide is sensitive to temperature and no  $N_2O$  emissions are formed in temperatures above 950 °C (Kilpinen, 2002b) or below 500 °C. Maximum  $N_2O$  production occurs in around 700 °C (IEA GHG R&D, 2000). In conventional combustion processes, temperatures are usually high, which results in negligible  $N_2O$  formation (but higher  $NO_x$ ) and subsequent destruction of formed  $N_2O$ . In fluidized bed combustion, which aims at low  $NO_x$  and  $SO_x$  emissions, nitrous oxide emissions can be significant because temperature in the combustion zone is usually between 800 and 950 °C. In this temperature range, 10–50% of fugitive cyano compounds from fuel nitrogen oxidise to  $N_2O$ . This share decreases with increasing temperature and is negligible at temperatures higher than 950 °C (Kilpinen, 2002b).

The effects of temperature and air ratio on  $N_2O$  emissions in specific cases are shown in Figure 1. Similar effects showing decrease in  $N_2O$  with increasing temperature, and increase in  $N_2O$  with increasing excess air, are reported also in various other studies, e.g. by Lu et al. (1994) and König et al. (1994). It should be noted that in FBC, amount of air used for combustion is larger than in conventional technologies (IEA GHG R&D, 2000). Low-temperature conditions, yielding high  $N_2O$  emissions, may also occur in some  $NO_x$  control processes and combustion of moist fuels, e.g. waste and sewage sludge (Kramlich & Linak, 1994). Therefore, drying of moist fuels may decrease  $N_2O$  emissions.

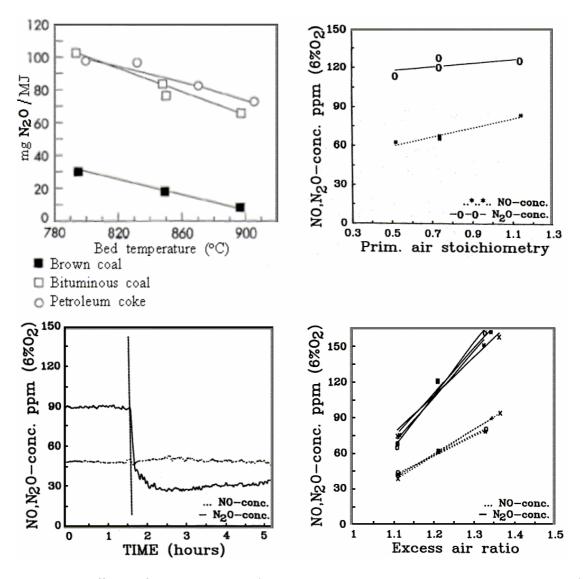


Figure 1. Effects of temperature and air ratio on  $N_2O$  emissions. Measurements with different bed temperatures were made in an 8 MW CFB boiler with air ratio 1.2 (Kilpinen, 2002b). The other measurements were carried out in a 12 MW CFB boiler. In the graph in the lower left corner, the drop of  $N_2O$  emissions is a response for leading 50% of the air supplied to the air register to the cyclone outlet duct (Åmand & Leckner, 1992).

In addition to the effect of lower temperature,  $NO_x$ -reduction measures like SNCR (Selective Non-Catalytic Reduction) may increase the amount of N<sub>2</sub>O via chemical reactions (Kilpinen, 1995; Korhonen et al., 2001). The N<sub>2</sub>O formation is more prevalent in urea- than in ammonia-based systems (Kilpinen, 1995; IIASA, 2004). When urea is injected to reduce NO emissions, typically more than 10% of the reduced NO is converted into N<sub>2</sub>O, whereas in the case of NH<sub>3</sub> injection this share is less than 5% (Kramlich & Linak, 1994). IIASA (2004) presents N<sub>2</sub>O emission factors for plants with SNCR that should be added to the emission factor for combustion device in question. These additional emission factors (8 mg N<sub>2</sub>O/MJ for NH<sub>3</sub> injection based SNCR and 25 mg N<sub>2</sub>O/MJ for urea based SNCR) are applicable for heavy fuel oil combustion (IIASA,

2004). But, there are also studies (e.g. Kilpinen & Hupa, 1998) concluding that SNCR does not necessarily increase  $N_2O$  emissions. Therefore, more measurements before and after SNCR of operating plant are needed for conclusions.

In selective catalytic reduction of  $NO_x$  (SCR), formation of  $N_2O$  depends on the catalyst type and operating conditions, but it is not estimated that use SCR leads to notably higher  $N_2O$  emissions (Kramlich & Linak, 1994). SCR may even reduce  $N_2O$  emissions. For example, according to the IPCC (1997), SCR reduces  $N_2O$  by 60%, at least when applied after gas firing systems or coke ovens. Some measurements indicate similar results also for other combustion devices with catalysers.

Air staging also affects  $N_2O$ , but the effect is rather complex. For example Hayhurst and Lawrence (1992) concluded that staged combustion reduces  $N_2O$ . In a study of König et al. (1994), it was shown that for one coal type (bituminous), emissions increased with increasing primary/secondary air ratio. But for other coal types studied, no notable effect was found. Kilpinen (2002b) reports possible decrease in  $N_2O$  emission with air staging. Low- $NO_x$ -burners may also have some influence on  $N_2O$  emissions.

When N<sub>2</sub>O is once formed, reactions breaking down N<sub>2</sub>O may also take place in the furnace. These are more efficient in high temperatures (Kilpinen, 2002b). For example in the freeboard of BFB it is common to find temperatures above 950 °C. These are due to combustion of volatiles and char in the freeboard. In addition, "hot spots" are more common with full load than with partial load. Part of N<sub>2</sub>O is destroyed in these "hot spots" even if the residence time is short. Increasing temperature may be the main reason if N<sub>2</sub>O emissions decrease when the load is increasing (Wihersaari & Palosuo, 2000). Correspondingly, the emissions may increase when the load is decreasing, also in other boilers than BFB (more information about partial load is available in Section 2.1.4). Residence time in the furnace might also be an important factor in some cases, for example in CFB where hot zones do not occur. Even in low temperatures some amount of N<sub>2</sub>O may be decomposed by reactions with hydrogen atoms (Kilpinen, 2002b).

In addition to combustion conditions, fuel type has obviously an effect on N<sub>2</sub>O emissions. The emissions of N<sub>2</sub>O often decrease with an increasing fuel-O/fuel-N ratio but absolute amount of N in fuel has also an influence on N<sub>2</sub>O emissions (Korhonen et al., 2001). Bituminous coals (mostly used coal type in Finland) and anthracite emit substantially more N<sub>2</sub>O than lower rank coals like brown coals (Lohuis et al., 1992; IPCC, 1997). In general, N<sub>2</sub>O emissions are larger for geologically older fuels, i.e. larger for coal than for peat, and larger for peat than for wood when other combustion conditions are equal (Kilpinen, 2002b). For example, it was shown in a study by Hiltunen et al. (1991), that N<sub>2</sub>O emissions from CFB boilers burning coal were 30–120 ppm, when they were 10–30 ppm when burning oil shale, peat and wood (IEA GHG R&D, 2000).

Studies of the role of pressure on N<sub>2</sub>O emissions have given contradictory results. The effect of pressure depends on other conditions in boiler, and is usually minor. Combustor type and size have also an impact on N<sub>2</sub>O. In a Japanese study including long measurement periods, N<sub>2</sub>O emissions from small-scale (laboratory scale) CFB were found to be 4–5 times higher that those from large scale CFB. It was supposed that the difference was due to stronger decomposition of N<sub>2</sub>O in larger boiler due to longer residence time (3–6 s vs. < 1 s). In large boilers, it is assumed that 50–80% of N<sub>2</sub>O formed in the bottom of the boiler is decomposed before it exits the boiler (Moritomi et al., 1999). Similar results were also obtained by König et al. (1994), who found out that emissions from pilot-scale boilers were an order of magnitude larger than those from larger boilers. Effect of residence time may be significant only for CFB boilers.

Studies of the effect of limestone feeding on  $N_2O$  emissions have also shown that the phenomenon is complex. Addition of CaO seems to lead to decreased  $N_2O$  emissions, but the reaction is affected by various other factors also (e.g. König et al., 1994; Hayhurst & Lawrence, 1992).

Formation and destruction of  $N_2O$  is a complex process that is not yet fully understood. In literature, some contradictory results are presented on, e.g. effect of CO in furnace on  $N_2O$  emissions. Some factors vary largely even in the same plant according to process conditions, and thus cannot be explicitly taken into account when developing emission factors for inventory purposes. The  $N_2O$  emission factors presented in literature vary significantly. In addition, partial load (discussed in Section 2.1.4) and unsteady situations have a great effect on  $N_2O$  emissions in some cases.

N<sub>2</sub>O emissions from fluidised bed combustion can be halved using specific measures in boilers. Options for FBC include increasing gas temperature in the upper part of the boiler over 950 °C by using, e.g. natural gas burning. The idea is to create a hot zone to the part of the boiler where fuel is totally combusted. Gas temperature should be over 950 °C for some milliseconds of time to decrease N<sub>2</sub>O emissions (Aho, 1997; Aho, 2004). For example in a study by Gustavsson (1994), effect of afterburning on N<sub>2</sub>O was studied in a 12 MW CFB boiler. Experiments revealed that increasing zone temperature by 150 °C reduced nearly all (90%) of N<sub>2</sub>O emissions. The effect was similar when secondary fuel was LPG, fuel oil, pulverised wood or pulverised coal. But, the amount of fuel needed to obtain a specific temperature varied by fuel type (Gustavsson, 1994). Even larger N<sub>2</sub>O reduction effect (up to 99%) was found by Marban et al. (1996) using methane and propane injection. This type of techniques have not yet been used in a commercial scale because N<sub>2</sub>O reduction has not been compulsory in many countries (Aho, 1997; Aho, 2004), but if such technologies were taken at use, emission factors should be changed respectively.

#### 2.1.2 Combustion technologies used in Finland

Many different fuels are used in stationary combustion in Finland. Coal and natural gas are the main fuels used in large scale energy production. The shares of peat, biomass and oil are also significant. Industrial power and heat production is relying heavily on biomass, especially on waste liquors (black liquor) and residual wood from pulp and paper industry. The main fuels in smaller scale boilers (residential and other) are oil and biomass. The share of each technology and fuel of CH<sub>4</sub> emissions from stationary combustion in Finland in 2002 is presented in Figure 2, and of N<sub>2</sub>O emissions in Figure 3, respectively. The figures are drawn by using emission data of 2002 inventory, and therefore they represent the knowledge of that time, i.e. emission factors recommended in this report have not been used. In addition, due to use of fuel mixes that is very typical for energy production in Finland, figures are only approximate. Use of fuel mixes are discussed more in Section 2.1.3.

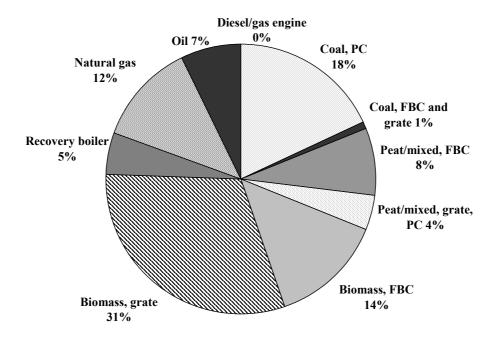


Figure 2. Share of different fuels and technologies of  $CH_4$  emissions from Finland (CRF 1A1 and 1A2) according to the 2002 GHG inventory.

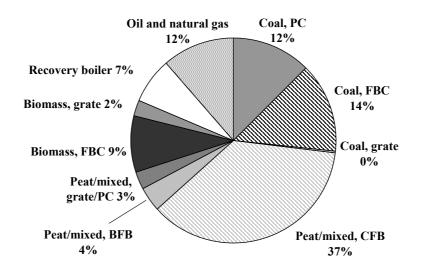


Figure 3. Share of different fuels and technologies of  $N_2O$  emissions from Finland (CRF 1A1 and 1A2) according to the 2002 GHG inventory.

Pulverised combustion is globally the most commonly used combustion technology for solid fuels (Kara et al., 2004). It is competitive in large installations, and has been used in Finland mainly in coal (Raiko et al., 2002) and also in few peat-fired boilers. In pulverised coal combustion, temperature scale in furnace is wide (Alvarez, 1990), peak temperatures being 2000 °C. Partial load may reduce the temperature, especially outside the flame region. Well-functioning pulverised combustion has very low N<sub>2</sub>O and CH<sub>4</sub> emissions. Japan is reporting negative CH<sub>4</sub> emissions for large pulverized coal burning plants as the measured CH<sub>4</sub> concentrations of the flue gases have been lower than the CH<sub>4</sub> concentrations in the air due to efficient oxidation in the boilers (GIO, 2004).

In Finland, fixed bed combustion (grates) is most commonly used for burning domestic solid fuels (i.e., peat and biomass) in small-scale combustion (< 5 MW). Grates have also been used in larger scale combustion, but this use is decreasing due to increasing use of fluidized bed combustion (Kara et al., 2004). CH<sub>4</sub> and N<sub>2</sub>O emissions from grates are usually low due to high combustion temperature (around 1500 °C in the combustion zone [Huhtinen et al., 1999]), but combustion of moist fuels may decrease temperature in the furnace. Because air ratio has a great effect on methane emission, also age and technology (e.g. combustion control and air ratio control equipment) of plant are important for emission factors. New grates are rarely built in Finland, and existing grate-firing plants are rather old. Therefore, average methane emission factors for grate firing plants are usually higher than for example factors for FBC plants.

Fluidized bed combustion (FBC) is continuously increasing its share of solid fuel combustion in Finland, while use of grates is decreasing. In Finland, a large variety of fuels and fuel mixes are combusted in fluidized beds: e.g. peat, biomass, coal and wastes. The technology is used typically in 5–200 MW<sub>e</sub> plants, but also larger plants

(400 MW<sub>e</sub>) are on the market (Kara et al., 1999; Hupa, 2004). In fluidized bed combustion, low-grade fuels or fuel mixes with varying particle size and moisture content can be combusted efficiently. Another advantage of fluidized bed combustion is possibility of in-process capture of SO<sub>x</sub>. Emissions of NO<sub>x</sub> are lower than in pulverised combustion due to low temperature (750–950 °C), and emission of CO and hydrocarbons are usually low due to good mixing (Kara et al., 2004). The residence time in FBC is usually long enough to allow complete oxidation of methane. The low temperature results in higher N<sub>2</sub>O emissions than in other combustion (CFBC). In bubbling fluidized bed combustion (BFBC) temperature peaks occur which decompose the formed N<sub>2</sub>O effectively, and the emissions are therefore lower. In pressurised fluidized bed combustion (PFBC), which is used to achieve better efficiency, the temperature is somewhat higher, about 900–1000 °C at full load (Kilpinen, 2002b). In Finland, PFBC has only been used in pilot plants.

In Finland, oil is most commonly combusted by burners in different kind of boilers. The temperature in oil burners is typically high, and therefore, e.g.  $N_2O$  and  $CH_4$  emissions are low (Kilpinen, 2002b). In unsteady situations temperature may decrease and emissions increase. Effects and occurrence of these situations need further research.

Gas turbines use mainly natural gas and oil as fuels in installations typically from 5 to 250 MW<sub>e</sub> in electricity production. Natural gas driven gas turbines with heat recovery (typically 5–50 MW) and GTCCs are usually used for combined heat and power production. Gas turbines used to cover peak load usually use oil as main fuel. In gas turbines, the temperature of the flue gas is usually 900–1400 °C (Kara et al., 2004), and combustion properties of natural gas enable complete combustion. Therefore N<sub>2</sub>O and CH<sub>4</sub> emissions are very low for natural gas combustion. For oil combustion in gas turbines, higher share of unsteady situations and properties of oil increase emissions.

Gasification of fuels aims at increasing the energy efficiency and reducing pollutants. Gasification techniques vary from biogas combustion in small-scale diesel engines (< 1 MW<sub>e</sub>) to large-scale Integrated Gasification Combined Cycle (IGCC) and fluidized beds (up to 200 MW<sub>e</sub>). The range of fuels gasified is also very large: e.g. wastes, wood and peat (Kara et al., 2004). There is very little data available on the CH<sub>4</sub> and N<sub>2</sub>O emissions from gasification. Emissions are calculated based on combustion of the product gas, whose composition may vary especially in the case of waste.

Large diesel and otto-cycle engines are used e.g. for combined heat and power production (CHP) and as peak power plants (Kara et al., 2004). The most typical fuels are heavy fuel oil and gasoil, but the use of natural gas and some other gases is increasing. There is very little data available on the  $CH_4$  and  $N_2O$  emissions from these

sources but according to experts, methane emissions from gas firing engines can be very high when part of the natural gas leaves the engines unburned, whereas  $N_2O$  emissions are assumed small. Both emissions (CH<sub>4</sub> and  $N_2O$ ) from oil firing seem to be small, at least in steady-state.

Temperatures are usually 1200–1300 °C in lime kilns, 1400–1600 °C in cement kilns and 1000–1100 °C in brick kilns (Hongisto, 2004). Due to high temperatures,  $CH_4$  and  $N_2O$  emissions are usually small.

Other combustion processes of importance are covered under the sections addressing the emissions of the specific fuels and greenhouse gases.

#### 2.1.3 Co-firing

In Finland, co-firing of different types of fuels is very common in, e.g. fluidized beds. Recent trends show increase in partial replacement of coal with biofuels and wastederived fuels (Kilpinen et al., 2003). It is estimated that unexpected dependencies may occur in co-firing, leading to significant changes in emission generation (e.g.  $N_2O$ ) when compared with combustion of one fuel type only (Kilpinen et al., 2003; Hupa, 2004). The influence of co-firing on the non-CO<sub>2</sub> emissions is estimated to be more important for  $N_2O$  than for CH<sub>4</sub> emissions because  $N_2O$  emissions are more depended on fuel type especially in FBC which is a typical co-firing technology.

For example in the study of Leckner et al. (2004), in co-combustion of coal and dried sewage sludge in CFB boiler, CO emissions decreased with increasing fraction of sludge, whereas  $N_2O$  emissions increased (N-content of sludge may in some cases be very high). No notable effect was found for the commonly used share of sludge (< 20% of energy) and the effect remained small even when share of sludge of energy was 50%. When sludge was wet, higher  $N_2O$  emissions occurred. In co-combustion of wood and sludge, dependency of the share of sludge and CO emissions was not so clear, but  $N_2O$  emissions increased with increasing share of sludge (N<sub>2</sub>O emissions from pure wood combustion were small). When coal and wood were compared as base fuels, both CO and N<sub>2</sub>O emission were lower with wood.

Shen et al. (2003) studied co-firing of coal and biomass in an experimental small bubbling fluidized bed. They concluded that for high-nitrogen content coal, co-firing of coal and biomass decreased the emissions of  $N_2O$  compared to pure coal combustion. The decrease in the emissions depended on the combustion temperature and the share of wood in the fuel as well as on the type of coal and biomass co-combusted. Large reductions in  $N_2O$  emissions were reported, e.g., for co-combustion of coal and wood at

temperatures around 950–975 °C and a wood content of 25%. The N<sub>2</sub>O emissions were approximately 40% lower for the fuel mix (120 ppm) than for coal combustion without any wood (220 ppm). Gulyurtly et al. (1994) also concluded that in co-combustion of coal and biomass, the level of N<sub>2</sub>O emissions decreased with increasing share of biomass. The decrease was larger than could be assumed by examining the fuel-N ratios only (Gulyurtly et al., 1994).

Hupa's (2004) presentation on the dependency of the  $N_2O$  emissions on the share of wood and coal in co-firing of the fuels shows similar results (see Figure 4). The presentation is based on tests in a 12 MW CFB boiler with coal, wood and sludge at Chalmers University in Sweden.

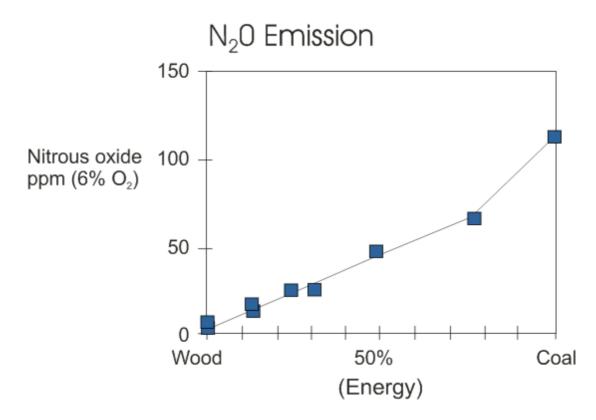


Figure 4.  $N_2O$  emissions as a function of the shares of wood and coal in co-firing of the fuels (12 MW CFB, constant temperature and air distribution) (Hupa, 2004).

Co-firing of peat and wood is common in Finland. But, there is not enough measurement data for establishing a relationship between shares of different fuels (wood and peat) and N<sub>2</sub>O emissions. According to the measurements in a 299 MW<sub>th</sub> CFB boiler the N<sub>2</sub>O emissions from burning peat were below the detection limit (2 mg/MJ) and for a fuel mix of peat (> 80%) and wood the N<sub>2</sub>O emissions were 4 mg/MJ. (Appendix B). Increase in N<sub>2</sub>O emissions with increasing share of wood is not a typical result and in this case it may be due to lower combustion temperature. Available emission factors for co-firing from literature are presented in Section 2.2.

In the case of methane, it has been noted that in co-combustion of wastes with other fuels, inclusion of waste (up to 26%) did not increase emissions of hydrocarbons. However, emissions may increase if fuel stocking changes due to variation in fuel size and energy content, which holds for all solid fuels (Huotari, 1996). Emission measurements done in laboratory-scale CFB for pelletized mix of different types of waste and peat, emissions of hydrocarbons were very low (under detection limit in some cases) (Vesterinen et al., 1996).

An approach to estimate the  $N_2O$  and  $CH_4$  emissions from co-firing could be to calculate the emissions assuming a linear relationship. This assumption would be applicable only in situations where the combustion conditions would otherwise be similar (e.g. combustion temperature or air ratio does not change remarkably). For inventory purposes, this may be the best available assumption because instantaneous share of one fuel type can vary between 0 and 100% but only annual shares of fuels combusted can be used for inventory purposes. Equation for calculating the emissions of co-firing is

$$Emissions = m_1 \cdot LHV_1 \cdot EF_1 + m_2 \cdot LHV_2 \cdot EF_2 + \dots + m_n \cdot LHV_n \cdot EF_n , \qquad (1)$$

where  $m_i$  is a mass of combusted fuel [t/a], LHV<sub>i</sub> is a lower heating value of fuel [MJ/t] and EF<sub>i</sub> is an emission factor [mg/MJ] of the fuel (given in Chapter 2.3).

The average emission factor for co-firing plant can be calculated by equation (2)

$$EF_{avg} = a \cdot EF_1 + b \cdot EF_2 + \dots + x \cdot EF_n , \qquad (2)$$

where a, b and x are shares of fuels [% of energy input] and  $EF_i$  is an emission factor. In most of the combustion plants in Finland, more than one fuel is combusted during the year. If the share of the main fuel is more than 80% of the annual energy input, the recommended emission factors in Chapter 2.3 give sufficiently accurate results.

In some furnaces, there are more than one combustion technology which use is remarkable (> 20% of energy). In this case, emission factors for both technologies could be used. For example, in the case of bark combustion in BFB complemented with oil firing by burner above that bed, emissions could be calculated by using both emission factor for bark combustion in BFB and oil firing by burner.

#### 2.1.4 Partial load

Partial load occurs rather often in boilers producing heat and/or steam, and to some extent in all boilers. Effect of load on GHG emissions depends on combustion technology and gas emitted. The effect is most important for N<sub>2</sub>O from FBC where temperature is low even at full load and decreases with decreasing load. In addition, effect is larger for CFB than for BFB because there are no "hot spots" that could destroy N<sub>2</sub>O in the CFB furnace due to circulation. Effect of partial load on temperature in the furnace and thus on N<sub>2</sub>O emissions of FBC is reported e.g. by Lee et al. (1994). In addition, measurement data presented in Appendix B shows that partial load increases N<sub>2</sub>O emissions from CFB. Effect of load on CH<sub>4</sub> emissions is unclear, but it seems that it is much smaller than the effect on N<sub>2</sub>O.

Load changes can be assumed most common for district heating plants (including CHP), because these plants are adjusted according to the need of heat. Notable load changes occur also in industrial CHP plants and more rarely in condensing power plants.

In Finland, district heating is a very common heating technology. Heat, steam and CHP production in district heating and industry covers over 80% of stationary combustion in Finland (Statistics Finland, 2003). Figure 5 presents the share of different fuels used in district heating, and Figure 6 fuels used in industry. Most common combustion technologies installed in district heating and industrial plants are presented in Figure 7.

Due to decreasing temperature in partial load, N<sub>2</sub>O emissions from these plants may be higher especially in the case of FBC (17% of industrial and district heating capacity). Production data obtained from one typical CHP plant show that even 60% of yearly production may be produced with load smaller than 75% and over 20% of energy may be produced with load < 50%. In Figure 8, production data of Pihlava CHP plant is presented as an example. It is a CFB plant burning peat and biomass as main fuels but also a notable amount of oil. In many CHP plants share of partial load may be higher.

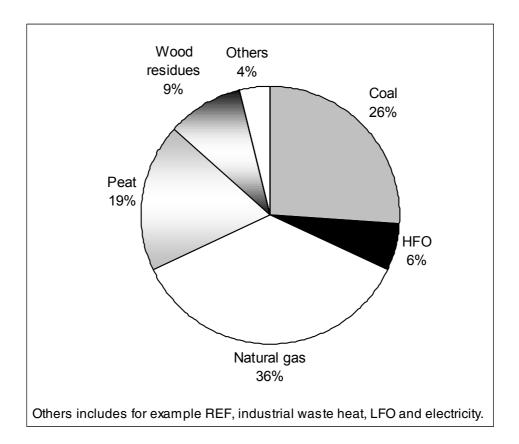


Figure 5. Share of different fuels [% of energy] used in district heat and CHP production in Finland in 2002 (Statistics Finland, 2003).

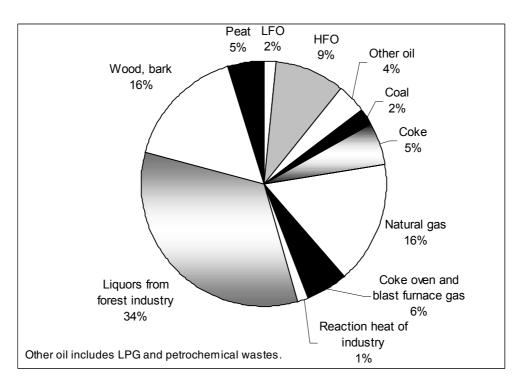


Figure 6. Share of different energy sources [% of energy] used in heat, steam and CHP production in industry in Finland in 2002 (Statistics Finland, 2003).

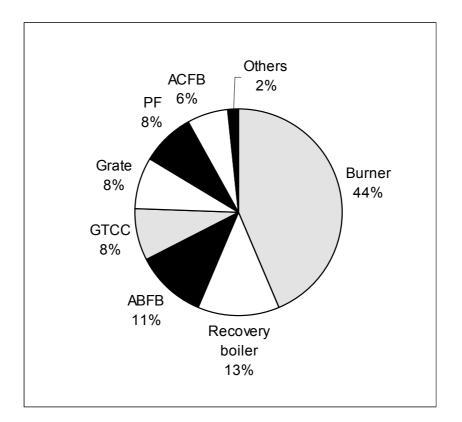


Figure 7. Share of different technologies of installed heat, steam and CHP production capacity [MW] in Finland in industry and district heating in 2002.

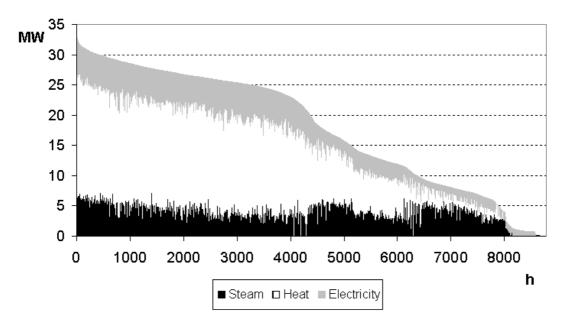


Figure 8. Duration curve for electricity, steam and district heat production from a typical Finnish CHP plant (Pihlava).

Because  $N_2O$  emission factors are different for partial load and full load and share of partial load may be notable, use of emission factors measured with full load may significantly underestimate emissions in the case of FBC operating frequently with partial load. Simple example is one peat firing CFB where emission factor of 26 mg/MJ was measured with 75% load (see Appendix B). Typical emission factor at full load for peat firing CFB boilers is < 2 mg/MJ. Using factor 26 mg/MJ for lower than 75% load and calculating emissions using production data presented in Figure 8, the resulting average annual emission factor is 11.7 mg/MJ.

The calculation presented above may be rather optimistic, because emissions may still increase when load decreases below 75%. For example, an emission factor of 36 mg/MJ was measured in one peat firing CFB at 65% load.

It is evident that partial load should be taken into account when developing  $N_2O$  emission factors. But, to be able to develop representative emission factors for the whole country, both measurement data from plants operating at partial load and data on occurrence of partial load would be needed.

It is likely that partial load does not have large effect on  $N_2O$  from other technologies than FBC. For example, Matsuda et al. (1994) studied formation of  $N_2O$  in pulverised coal combustion using a bench scale combustor. In partial load (60%), rather high  $N_2O$  levels (> 20 ppm) occurred awhile in the combustion area. This was due to lower temperature and higher oxygen concentration than in full load. But, the concentration decreased due to  $N_2O$  destruction in the furnace, and was < 3 ppm in the outlet.

In the case of CH<sub>4</sub> emissions, air ratio and mixing are more relevant factors than effect of temperature. Thus there is no evidence to give different CH<sub>4</sub> emission factors for power plants according to different loading.

#### 2.1.5 Measurement techniques

Concentration of  $CH_4$  and  $N_2O$  is usually rather small in flue gases from combustion. Because these emissions are often close to the detection limit of the measurement instrument, technique used in measurements should be given attention when evaluating measurement results.

In the turn of the 1990s nitrous oxide ( $N_2O$ ) was measured usually by non-continuous methods, such as bag sampling followed by an analysis with gas chromatography (GC). This method, however, was prone to errors. It was noticed that too high  $N_2O$  concentrations were determined in bag samples because NO, SO<sub>2</sub> and H<sub>2</sub>O formed N<sub>2</sub>O

as an artifact. Therefore, based on these measurements, wrong conclusions were drawn about nitrous oxide emissions.<sup>4</sup>

Uncertainty in measurement instruments, when used correctly, is often rather low. But, the total uncertainty of measurements may be substantially higher, due to, e.g. possible human errors, errors in calibration, interpretation or results etc. According to Cooper (2001) the estimated or instrument manufacturer specified uncertainty of GC measurements for  $CH_4$  and  $N_2O$  was  $\pm 5\%$  (measurement results in ppm), but the total uncertainty in measurement results was estimated at  $\pm 12\%$ .

The development of new techniques, such as FTIR technique (Fourier transform infrared spectroscopy), NDIR (non-dispersive infrared technique) and GFC-IR (gas filter correlation infrared technique) have enabled continuous  $N_2O$  measurements. Traditionally, FTIR has been used in laboratory applications, but development in e.g. interferometers has made it possible to use the method also in field applications. FTIR is based on the different wavelengths that compounds of the flue gases absorb, because most compounds have a specific wavelength. With interferometer, an interferogram can be obtained from IR-beam. Interferometer typically forms from stationary and moving mirrors. Interferogram is converted to simpler form by Fourier transform and result is a spectrum where concentrations can be calculated. The detection limit of FTIR depends on methods used for data improvement but is around 1 ppm for most compounds (corresponding approximately to a value of 0.5 mg N<sub>2</sub>O/MJ).

The measurement of  $CH_4$  was earlier based on assumptions on the share of methane of TOC-concentration that was measured using FID-technique. These assumptions were not always correct.  $CH_4$  emissions can nowadays be measured directly using FID-technique. According to some studies, properties of the catalyst needed to separate methane from other TOC in FID measurements might change during the measurement and lead to erroneous results. FTIR-technique seems to be the most reliable way to measure also methane concentrations continuously. But, IR-technique based methods have some difficulties due to the cross-interferences of other components. For more information on VOC measurements, see Appendix E.

In studies of Fabritius et al. (2002) and Korhonen et al. (2001)  $N_2O$  was measured with Servomex Xentra 4900 analyser, based on infrared absorption which was monitored at characterised wavelength and gas filter correlation technique (NDIR). The reported

<sup>&</sup>lt;sup>4</sup> These erroneous results have neither been used when compiling emission factors for the first Finnish GHG inventories, in compilation of the IPCC emission factors, or any other emission factors presented in this report.

confidence limit was 2 ppm. Also two different methods (NDIR and FTIR) were compared in one plant and the  $N_2O$  results were almost identical (Fabritius et al., 2002).

In the case of TOC measurements, Fabritius et al. (2002) used Signal Model 3000 analyser, based on the flame ionisation detector (FID). The sample gas was conducted via heated particle filter and a heated sample line (150 °C) to the detector which consisted of a combustion chamber and a burner. The organic compounds were ionised to electrons and positive ions in the hydrogen/helium flame. The ions and electrons were collected using the collector electrode placed near the flame. The ionisation current between the collector electrode and burner nozzle was converted to the measurement signal. The reported confidence limit for the measurements was 2 ppm. NMVOC concentrations can also be measured with FID, but a catalyst is needed. In Fabritius et al. (2002) only "total mode" of FID was used. However, most of the results from this source are valid also for methane, because TOC concentration was below the detection limit (if TOC emission is < 1 mg/MJ also CH<sub>4</sub> is < 1 mg/MJ).

In Fabritius et al. (2002)  $NO_x$  was measured with chemiluminescense based Monitor Labs 9841A analyser.  $SO_2$  was measured with UV-fluorescence based Monitor Labs 9850 analyser.  $NO_x$  and  $SO_2$  analysers were connected to dilution probe EPM 797. The reported confidence limit for these measurements was 5 ppm.  $O_2$  was measured using oxygen paramagnetism based M&C PMA 25 analyser. CO and  $CO_2$  were measured with infrared absorption based Hartmann & Braun Uras 10 E analyser (Fabritius et al., 2002).

In Fabritius et al. (2002)  $N_2O$ ,  $O_2$  and CO analysers were connected with Teflon sample lines to sample gas cooler Hartmann & Braun SCC, which removed the moisture from the sample gas flow. TOC,  $NO_x$  and  $SO_2$  results were given on a wet basis (Fabritius et al., 2002).

#### 2.2 Emission factors from literature

 $N_2O$  and  $CH_4$  emission factors used in the national greenhouse gas inventory of Finland as well as emission factors from different literature sources, research and measurements are presented in the following sections.<sup>5</sup> All emission factors are given as [mg/MJ<sub>fuel</sub>] if not otherwise stated.

Two main types of data on the emission factors can be found: (i) primary sources which are based on measurements and/or theoretical modelling and estimates of the emission

 $<sup>^{5}</sup>$  A summary of the CH<sub>4</sub> and N<sub>2</sub>O emission factors used in the Finnish inventory is presented also in Appendix A. Publicly available data from measurements are compiled in Appendix B, and emission factors used by other countries into Appendix C.

factors, and (ii) secondary sources, which are summarising, analysing and/or using data originating from other sources.

There is very little primary data publicly available on the  $N_2O$  and  $CH_4$  emission factors. Primary sources used in this study comprise scientific literature and published and unpublished reports on measured data on the emissions. When estimating emission factors for GHG inventories which represent annual emissions, all disturbances and unsteady situations should be included in the estimates. This should be taken into account when comparing measurement results with steady load and emission factors from secondary sources. In addition, some plants may not have as good automation or control devices as measured plants, and this should also be taken into account in emission factors. For example, poor air ratio control may result hundred times higher methane emissions than emissions from similar plant with good control devices.

The main secondary data sources used are the Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 1997),<sup>6</sup> the IPCC Emission Factor Database (IPCC, 2004), the Joint EMEP/Corinair Atmospheric Emission Inventory Guidebook (Corinair, 2004) and a report by the Finnish Environment Agency on guidance on reporting of emissions from energy production to national registries (SYKE, 2004). The report of SYKE (2004) was based on currently used emission factors and findings of a working group aiming at improving current emission factors.

AP-42 is a compilation of air pollutant emission factors made by United States Environmental Protection Agency. The emission factors in AP-42 are based on HHV of fuel and are therefore not presented in the tables of this Section. In a study where emission factors from IPCC (1997) were transformed to HHV-based emission factors, the CH<sub>4</sub> emission factors from AP-42 diverged only less than  $\pm 15\%$  (all less than 0.3 mg/MJ) from the IPCC values for coal and oil but for natural gas combustion emission factors from AP-42 were 91% greater for boilers and 16% greater for engines than factors from IPCC (1997). For gas turbines AP-42 factor for CH<sub>4</sub> was 46% smaller than factor from IPCC (1997). For combustion of wood residues AP-42 presented significantly smaller CH<sub>4</sub> emission factor than IPCC (URS Corporation, 2003; AP-42, 2005).

Emission factors for N<sub>2</sub>O from bituminous coal combustion using other technologies than FBC are small (< 2 mg/MJ) according to both sources (IPCC, 1997; URS Corporation, 2003). For FBC the AP-42 has almost 60% smaller factors than the IPCC (1997). For lignite combustion in fluidized beds and for oil combustion in boilers the N<sub>2</sub>O emission factors are almost equal. IPCC (1997) does not present  $CH_4$  or  $N_2O$ 

 $<sup>^{6}</sup>$  The IPCC Good Practice Guidance for National Greenhouse Gas Inventories (IPCC, 2000) does not provide any additional information on emission factors for CH<sub>4</sub> and N<sub>2</sub>O.

emission factors for oil combustion in gas turbines but AP-42 has the same factors for oil combustion in turbines and in boilers (URS Corporation, 2003). IPCC (1997) also presents "not available" for N<sub>2</sub>O emissions from natural gas or wood combustion but AP-42 gives factors 0.9-1.3 mg/MJ for natural gas and 5.6 for combustion of wood residues. These factors are based on HHV, for LHV factors are some percents higher (URS Corporation, 2003).

Emission factors used by other countries in their reporting to the UNFCCC are collected into Appendix C. These emission factors are partly primary emission factors, and partly so-called implied emission factors (IEFs) calculated from the emissions and activity data reported for a specific IPCC category, and therefore representing aggregated data. The basis of the aggregation may differ from country to country and cannot usually be specified from the data available. The emission factor data collected from these sources is used to estimate the range of emission factors used in inventory compilation, and also to assess how the emissions factors used in the preparation of the Finnish GHG inventory compare to this range. In Germany, a detailed study of emission factors for small-scale combustion was carried out in 2000 (Pfeiffer et al., 2000), and these results are also presented in Appendix C.

In this Section (2.2) the collected data on the emission factors are presented by greenhouse gas, fuel and technology and analysed to find the best estimates for the Finnish conditions. Fuels are divided to eight sections (Sections 2.2.1-2.2.8), which mainly follows the classification of IPCC. For example, factors from literature for refined fuels are presented in the section of the original fuel e.g. coke (including petroleum coke), coke oven gases and blast furnace gas are presented in the Section 2.2.1 Coal. Products of gasification are presented in Sections covering the fuel gasified. Exceptions include wastes which form one section. Wood residues and woodwaste are presented in Sections Biomass and Waste, respectively, but the classification depends also on fuel type and literature source. Two last sections are for combustion processes where there is no main fuel (share of each fuel is < 50%), fuel is not specified or emissions are more process than fuel depended (e.g. lime kilns). The evaluation of suitability of the emission factors to conditions at Finnish plants is based on the data collected from the various sources, and discussions with combustion and measurement experts. The validity of the emission factors during the whole time series is assessed based on data on the changes in fuel use, technology and the way the facilities have been operated during the period of interest. But, due to lack of quantitative data on changes in emissions in time series, no exact recommendations for different emission factors for different years are given. The evaluation of the emission factors used in the Finnish GHG inventory is summarised in Section 2.3 accompanied with recommendations for changes.

Need for further investigations (measurements and data collection) to increase knowledge of the emission factors is estimated in Chapter 3 taking the importance of the emissions in the Finnish inventory into account.

#### 2.2.1 Coal

Solid fuel combustion, comprising in addition to coal also coke oven and blast furnace gas in inventory (but not wood and peat), covered 14% of methane emissions and 25% of N<sub>2</sub>O emissions from stationary point sources in Finland in 2002 (20% and 29% in 1990). Over 99% of coal is used in large installations (CRF categories 1A1 and 1A2). Coal is combusted in Finland using pulverised combustion, grates and fluidized beds. Pulverised combustion is the most important technology for coal combustion in power production. Use of grates has decreased since 1990.

#### 2.2.1.1 CH<sub>4</sub> from point sources (coal)

The implied average emission factor for  $CH_4$  from solid fuel combustion (3.6 mg  $CH_4/MJ$ ) has not changed remarkably between the two years (1990 and 2002). The implied emission factor is smaller for coal than for the other fuels. This is in agreement with the known fact that combustion in large installations is nearly complete. A high emission factor is given for small (< 1 MW) circulating fluidized beds burning coal as the main fuel.

The  $CH_4$  emission factors used in the Finnish 2002 GHG inventory for coal combustion are given in Table 1. For coke ovens and blast furnaces, emission factor of 0 mg/MJ is used (see Section 2.2.8).

Table 1.  $CH_4$  emission factors [mg/MJ] for coal combustion (and co-firing when coal is the main fuel) used in national GHG inventory of Finland.

Fuel Technology/Size [MW]		<u>&lt;</u> 1	1–5	5–15	15
Coal 50-100%	CFB with air staging				
Coal	Coal CFB without air staging			5	4
Coal	BFB + flue gas recirculation				5
Coal	Burners, grate, burner + grate, others		7	4	4
Coal	Grate + flue gas recirculation		8		
Coal 50-80%	PFBC, grate			4	4
Coal 50-80%	No specification			3	8

For coal combustion,  $CH_4$  emission factors from different literature sources varied from 0.2 to 14 mg  $CH_4/MJ$  in large scale combustion (Table 2). The emission factors in the measurements are typically very low, and often below the detection limit (about 1 mg  $CH_4/MJ$  in most measurements used in this report).

Table 2. Emission factors [mg CH<sub>4</sub> /MJ] given in literature for coal, coke, coke oven gas and blast furnace gas combustion.

		Corinair, 2004 <sup>(3</sup>		SYKE, 2004 <sup>(1</sup>	IPCC, 1997 <sup>(2</sup>	Measured data (Appendix B)
Fuel	Technology	Utility	Industrial	>15 MW		~ 400 MW
Coal	Pulverized firing			1	0.7 <sup>(4</sup> 0.9 <sup>(5</sup>	< 1
Coal	FBC/PFBC	0.6	2.4	1	1.0	
Coal	Cyclone furnace				0.2	
Coal	Coke oven				1	
Coal	Grate/stoker firing	0.7		4	1.0 14 <sup>(6</sup>	
Coal	Hand-fed units				87	
Coal	No specif. < 50 MW		-511			
Coke	No specification		2–15 <sup>(7</sup>			
Petroleum coke	No specification		1.5 <sup>(7</sup>			
Coke / petroleum	No specif. < 50 MW	1	5-200			
coke						
Coke oven gas	No specification	0.	.3–4 <sup>(7</sup>			
Coke oven gas	Gas turbine < 50 MW		2			
Coke oven gas	No specif. < 50 MW		02–12			
Blast furnace gas	No specification	0.3–2.5 <sup>(7</sup>				
Blast furnace gas	No specif. < 50 MW	0.02–4				

 $^{1} > 50\%$  coal.

<sup>2</sup> For bituminous and sub-bituminous coal in utility/industrial scale.

 $^{3}$  > 50 MW unless otherwise stated.

<sup>4</sup> Dry bottom boilers.

<sup>5</sup> Wet bottom boilers.

<sup>6</sup> Underfeed stokers.

<sup>7</sup> In Corinair (1990) as presented in Corinair (2004).

Emission factors from other countries presented in Appendix C for methane emissions from public power plants combusting coal vary from 0.2 to 5 mg/MJ. Denmark gives a rather large EF for < 300 MW district heating plants (15 mg/MJ). IEFs for stationary combustion in industry are higher, 0.3–200 mg CH<sub>4</sub>/MJ. The variation between countries is large, and because reporting is made at rather aggregated level, emission factors cannot be compared by technology and size.

Emission factors in literature and measurements indicate that the emission factors used in the Finnish inventory are at the higher end of the range. For pulverised coal combustion and FBC, an emission factor of 1 mg CH<sub>4</sub>/MJ would produce more accurate estimates for > 5 MW plants. The use of a small emission factor is also backed up by theoretical arguments: in PF temperatures are high enough, and in FBC the residence time is long enough to prevent methane emissions in stable conditions. The real emissions could be even smaller than 1 mg CH<sub>4</sub>/MJ in modern units, as some measured emissions were below the detection limit (1 ppm). This gives also support for the Japanese estimate (GIO, 2004) that efficient combustion could even reduce the methane concentration in the atmosphere. Current methane concentration in atmosphere is about 1.7 ppm<sub>vol</sub> (IPCC, 2001) – which is above the detection limit. But, it has to be noted that detection limit depends on many factors and is not the same for all measurements.

In smaller and older plants control and automation devices might be less advanced and share of unsteady situations is likely to be higher. There is very little information on emissions from small boilers (< 5 MW), but it is assumed that emissions are higher (4 mg/MJ) than in large boilers mainly due to shorter residence time and other above mentioned reasons. Emission factor for grate firing could be somewhat larger in < 50 MW units (4 mg/MJ) than in > 50 MW units (1 mg CH<sub>4</sub>/MJ). For coke oven gas and blast furnace gas, emission factors recommended for gas combustion (Table 36) can be used.

#### 2.2.1.2 N<sub>2</sub>O from point sources (coal)

The implied average emission factor for  $N_2O$  from solid fuel combustion (3.7 mg  $N_2O/MJ$ ) was the same in 1990 and 2002. The emission factors used in the Finnish inventory are given in Table 3.  $N_2O$  emission factor is high for fluidised bed combustion (70 mg  $N_2O/MJ$ ), but lower for other technologies (2–4 mg/MJ). For coke ovens and blast furnaces, emission factor of 2 mg/MJ is used.

Table 3.  $N_2O$  emission factors for coal combustion used in the national GHG inventory of Finland [mg  $N_2O/MJ$ ].

Fuel	Technology	<b>Emission factor</b>		
Coal	Grate, burner + grate	4		
Coal 50-80%	Grate	2		
Coal 50-100%	FBC	70		
Coal 50-100%	PFBC, burners, other boilers	2		

Emission factors from other literature sources are compiled in Table 4. Nitrous oxide emission factors for coal combustion vary from 0.5 to 96 mg  $N_2O/MJ$  depending on combustion technology.

Fuel	Technology	SYKE, 2004	IIASA, 2004	<b>Corinair, 2004</b> <sup>(1)</sup>	IPCC, 1997 <sup>(2</sup>	Measurements (Appendix B)
Coal	PF	1	1.4	0.8	0.5-1.6 (3	< 1-1.7
Coal	FBC	70	80 <sup>(4</sup>		96 <sup>(5</sup>	14–37 (6
Coal	Grate	4		0.8	1.6	
Coal	Other stoker boilers			0.8	1.6	
Coal	Cyclone furnace		1.4	0.8	1.6	
Coal	Hand-fed units				1.6	
Coal	Coke oven			0.8		
Coal	No specif. < 50 MW			5-30		
Coke	No specification			1.4–25 <sup>(7</sup>		
Petroleum coke	No specification			14 <sup>(7</sup>		
Coke, petroleum coke	No specif. < 50 MW			1.4–14		
Coke oven / Blast furnace gas	No specification			1.1–3 <sup>(7</sup>		
Coke oven / Blast furnace gas	Gas turbine < 50 MW			3		
Coke oven gas	No specif. < 50 MW			1–12		
Blast furnace gas	No specif. < 50 MW			0.8-34.6		

Table 4.  $N_2O$  emission factors for coal, coke, coke oven gas and blast furnace gas combustion [mg/MJ] from literature.

<sup>1</sup> 50–300 MW if not otherwise stated.

<sup>2</sup> Bituminous and sub-bituminous coal.

<sup>3</sup>0.5 for dry bottom tangentially fired plants and 1.6 for other PF.

<sup>4</sup>IIASA gives the same EF for all fuels in CFB.

<sup>5</sup> 42 mg/MJ for lignite.

<sup>6</sup> 14 mg/MJ is for lignite. Ahlstrom presents a scale of 15–60 mg  $N_2O/MJ$  for coal combustion in CFB (Korhonen et al., 2001). For PFBC the scale is 8–18 mg/MJ (Appendix B).

<sup>7</sup> Presented as Corinair90 data in Corinair (2004).

In the Corinair 1990, emission factors varied from 2.5 to 100 mg  $N_2O/MJ$ . For petroleum coke and coking coal a specific factor of 14 mg  $N_2O/MJ$  was given (Corinair, 2004).

 $N_2O$  IEFs from other countries (Appendix C) show variation from 0.9 to 20 mg/MJ for public and industrial power plants. For example in Germany, where emission factors were available by technology, emission factor of 4 mg/MJ is used for all other technologies than for FBC, for which emission factor is 20 mg/MJ (emission factors for brown coal were lower).

Most measured values for pulverised coal combustion presented in Appendix B were below 1 mg/MJ in stable conditions. This and the values in Table 4 indicate that 1 mg  $N_2O/MJ$  would be the best estimate for the emission factor for pulverized coal combustion when unsteady situations are included.

Data from literature and measurements indicate that for CFB the emission factor would be in the range of 30–80 mg/MJ. It is supposed that an emission factor of 30 mg/MJ could be used for CFB-plants with relatively rare occurrence of partial load (other than district heating), and 60 mg/MJ for district heating CFB-boilers, where partial load plays a significant role. For BFB the emission factor should be lower (20 mg/MJ) due to "hot spots" that destroy N<sub>2</sub>O, even though many literature sources show rather high values.

For PFBC factors between 8-18 mg/MJ were measured from one boiler in Sweden in steady state. Other studied reviewed revealed that pressure did not have any large effect on N<sub>2</sub>O emissions, and therefore the same emission factors could be used for pressurised FBC as for FBC in atmospheric pressure.

For other combustion technologies (e.g. grates), an emission factor of 3 mg/MJ is suggested. For coke oven gas and blast furnace gas, emission factors given for combustion of other gases than natural gas could be used.

#### 2.2.1.3 Small-scale combustion of coal

The use of coal in small-scale applications is minor in Finland (0.5% of energy use in small-scale combustion in 1990, 0.1% in 2002). In Finnish greenhouse gas emission inventory, emission factor of 300 mg  $CH_4/MJ$  is used for coal combustion in small-scale applications in 2002. Table 5 presents emission factors from the IPCC (1997) and Corinair (2004).

<i>Table 5. CH</i> <sub>4</sub> <i>emission factors</i>	[mg/MJ] from literature	e for small-scale combustion of	of coal.
		<i>jet 2000 2000 000000000000000000000000000</i>	

	Corinair, 2004	IPCC, 1997
Commercial boilers	10	10
Residential anthracite space heaters		150
Coke, petroleum coke and coal briquettes	225	
Coke oven gas	2.5	
Other residential combustion of coal	450	

In other countries (Appendix C),  $CH_4$  emission factors for small-scale combustion of coal vary from 4 to 300 mg/MJ. It seems evident that emission factors are much larger (more than an order of magnitude) in households than in other small-scale applications. Therefore, emission factors used in Finland could be divided in different sub-categories according to size of the application. An emission factor of 250 mg/MJ could be used for households, and 10 mg/MJ for other small-scale combustion.

The N<sub>2</sub>O emission factor used in Finland for small-scale combustion was 2 mg/MJ in 1990 and 4 mg/MJ in 2002. IPCC and Corinair did not present N<sub>2</sub>O emission factors for residential coal combustion. Other countries (Appendix C) use emission factors from 1.4 to 20 mg/MJ. Based on this data, Finnish emission factors seem to be at the lower end of the range. Emission factor 6 mg/MJ could be a good estimate for the whole time series.

#### 2.2.2 Oil products

Liquid fuel combustion, which comprises all oil products, covered 14% of methane and 21% of N<sub>2</sub>O emissions from large scale combustion (point sources) in Finland in 2002.<sup>7</sup> The use of liquid fuels is distributed nearly equally between large (CRF categories 1A1 and 1A2) and small installations (CRF categories 1A4 and 1A5). Use of oil has decreased since 1990 due increased use of biomass and enlargement of district heating network. Increased use of natural gas has also replaced heavy fuel oil (Hoffman et al., 2004). Oil is usually combusted as main fuel or as auxiliary fuel using burners. A small share of oil is also used in gas turbines. Natural gas has increased its share also in these applications since 1990. In Finland, gasoil (light heating oil) is used in stationary diesel engines.

#### 2.2.2.1 CH<sub>4</sub> from point sources (oil products)

The average implied emission factor for  $CH_4$  from combustion of liquid fuels in point sources was somewhat higher in 1990 than in 2002 (6.7 mg  $CH_4/MJ$  vs. 6.3 mg  $CH_4/MJ$ ). Emission factors for oil combustion used in the Finnish inventory varied from 2 to 10 mg  $CH_4/MJ$  (Table 6). Emission factors found in literature ranged from 0 to 21 mg  $CH_4/MJ$  depending on oil type, combustion technology and literature source (Table 7). Measured values for combustion of heavy fuel oil (65 MW<sub>th</sub>) were below the detection limit (1 mg  $CH_4/MJ$ ), even under conditions where the boiler was operating at partial load (see Appendix B).

<sup>&</sup>lt;sup>7</sup> Nearly half of oil products combusted in Finland is used in transportation, but this report is concentrated on stationary emission sources.

Table 6.  $CH_4$  emission factors [mg/MJ] for oil combustion used in the national GHG inventory of Finland.

Fuel	Technology		Size class		
		≤1 MW	1–15 MW	15–50 MW	> 50 MW
Oil	Cyclone	_	_	10	—
Oil	BFB	—	5	—	—
Oil	Other boilers	10	8	8	8
Oil	Gas turbines	—	8	8	8
Oil 50-80%	All	-	8	3	4
Oil	Diesel engine	2	2	2	2

In other countries (Appendix C) IEFs for oil combustion vary from 0.1 to 3.5 mg/MJ for all oil types. IEFs are somewhat higher for residual oil than for gas oil and fuel oil.

Source			Corinair, 2004	ir, 200	4		1907, 1997	1997	SYKE, 2004 (>15 MW)	Measured data (Appendix B)	d data lix B)
Fuel	Residual oil	Gasoline	Diesel/ Gasoil	LPG	LPG Kerosene	Refinery gas	Distillate Residual fuel oil oil	Residual oil	All oil	LSMGO <sup>(1</sup> HFO	HFO
Utility boilers	$0.7^{(6)}$		$0.03^{(6)}$			0.02-2.5 (7	6.0	$0.9^{(3)}$	$1^{(2)}$		$\frac{1}{2}$
Industrial boilers	$2.9^{(6)}$						0.2	ю	$1^{(2)}$		
Gas turbines	1–3		1–21	1 <sup>(5</sup>	1(5	2 <sup>(5</sup>			8	1	
Stat. engines (> 50 MW)	ю		1.5			0.02–2.5 <sup>(7</sup>	All oil	All oil 4.0 <sup>(4</sup>	2		
Stat. engines (< 50 MW)	0-8	49	0-14		0.0-7.4				2	0–2	2–3
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Table 7. Emission factors given in literature for combustion of oil products [mg CH4/MJ].

<sup>1</sup> LSMGO = Low sulphur marine gas oil. Measurements are for main and auxiliary engines of ships. <sup>2</sup> For all fuels where oil content is > 50%.

Also for shale oil.

<sup>4</sup> > 0.5 MW utility engines. For industrial engines (> 0.5 MW) EF of 0.0 is given. <sup>5</sup> Industrial < 50 MW gas turbines.

<sup>6</sup> For >50 MW plants.

<sup>7</sup> Presented as Corinair90 data in Corinair (2004) without technical specification.

Table 8. Emission factors given in literature for combustion of oil products [mg  $N_2O/MJ$ ].

							Corinair, 2004	2004				Measured
Unit Size	Technology	SYKE, 2004	SYKE, IPCC, 1997 <sup>(1</sup> 2004	IIASA, 2004	Gasoline	Kerosene	Naphtha	RFO	Diesel/ Gasoil	Gasoline Kerosene Naphtha RFO Diesel/ Refinery LPG Gasoil gas	LPG	data (Appendix B)
	Burner	1			14		, ,			1.5	1 - 14	
< 50 [MW]	Gas turbine		0.3 (RFO and			14	71	2.5-25	2.5–25 0.5–25	3	14	6 (2
	Diesel/gas engine	2	shale oil)	0.6	2	2		1.1–2.1	1.1–2.1 0.6–14	1.5	1-14	0-7 (2
	Burner	1	0.4 (DFO)					46.5	46.5 15.7	J = 1Λ <sup>(3</sup> 7 Λ 3 <sup>(3</sup>	7 1 2 <sup>(3</sup>	
> 50 [MW]	Gas turbine					$14^{(3)}$	14 (3	2.5-14	2–3	2.J-14	2- <b>4</b> -7	
	Diesel/gas engine	2					ţ	2.5	2.5	2.5		
RFO = residua	RFO = residual fuel oil, DFO = distillate fuel oil.	te fuel oil.										

<sup>1</sup> Utility/industrial boilers. <sup>2</sup> Fuel in gas turbine and in most diesel engines is low sulphur marine gas oil. <sup>3</sup> From Corinair90 (presented in Corinair 2004) without technical specification.

In addition to factors presented in previous Tables, many emission factors from Corinair90 were presented in Corinair 2004. There was no technical specification for plants but factors for methane varied from 0.1 to 10 mg/MJ for residual fuel oil and from 0.1 to 8 for gasoil.

Measurements presented in Appendix B support an emission factor of 1 mg CH<sub>4</sub>/MJ for heavy oil combustion in large (> 50 MW) boilers. For engines (> 50 MW) a factor of 2 mg CH<sub>4</sub>/MJ is recommended due to more incomplete combustion. In smaller units, emission factors may be somewhat larger (3–4 mg/MJ) due to more incomplete combustion and relatively high occurrence of start-ups and shut-downs in peak and emergency power plants.

#### 2.2.2.2 N<sub>2</sub>O from point sources (oil products)

The average implied emission factor for  $N_2O$  from stationary combustion of liquid fuels was somewhat lower in 1990 than in 2002 (3.7 mg N<sub>2</sub>O/MJ vs. 5.7 mg N<sub>2</sub>O/MJ). The N<sub>2</sub>O emission factors used for oil combustion in the Finnish GHG inventory are given in Table 9. The N<sub>2</sub>O emission factor for oil combustion is low (1–2 mg N<sub>2</sub>O/MJ) for applications other than fluidised beds and diesel engines (around 30 mg N<sub>2</sub>O/MJ).

Table 9.  $N_2O$  emission factors for oil combustion used in the national GHG inventory of Finland.

Fuel	Technology	<b>Emission factor</b>
Oil	FBC	30
Oil 50–100%	Other	2
Oil	Gas turbine	1
Oil	Diesel engine	31

 $N_2O$  emission factors from literature are given in Table 8. For oil combustion,  $N_2O$  emission factors vary from 0.3 to 47 mg  $N_2O/MJ$ . For diesel engines, measured values vary from 0 (below detection limit) to 7 mg  $N_2O/MJ$ . In many of the measurements done by Cooper (2001) the load was low or the fuel combusted had a high nitrogen content. The impact of the varying conditions on the average factor was found to be small for diesel engines. In Ahlstrom pyroflow CFB boilers, 5–15 mg/MJ is reported as a typical  $N_2O$  emission factor in oilshale combustion (Korhonen et al., 2001).

In other countries (Appendix C)  $N_2O$  emission factors for oil combustion varied from 0.6 to 2 mg/MJ for fuel oil and gasoil (expect Ireland that reported higher values), and up to 5 mg/MJ for residual fuel oil.

According to measurement data (Appendix B) emission factor for oil combustion in boilers may be  $< 1 \text{ mg N}_2\text{O/MJ}$ . When disturbances are taken into account, 1 mg/MJ would be the best approximation. For gas turbines and engines emission factor should be somewhat higher (4 mg/MJ) due to shut-downs and start-ups. For FBC, suggested emission factors range from 20 to 30 mg/MJ according to combustion technology (BFB or CFB) and occurrence of partial load.

#### 2.2.2.3 Small-scale combustion of oil products

Oil combustion represented over half of energy used in small-scale combustion in Finland in 2002. CH<sub>4</sub> emission factor used in the Finnish GHG inventory is 10 mg CH<sub>4</sub>/MJ and that of N<sub>2</sub>O 2 mg/MJ. The emission factors are the same for LFO and HFO. Table 10 presents CH<sub>4</sub> emission factors from literature.

Table 10. CH<sub>4</sub> emission factors for small-scale combustion of oil [mg/MJ].

Source	Co	rinair, 2	004		IPCC,	1997	
Fuel	RFO <sup>(1</sup>	Gasoil	LPG	LPG	DFO <sup>(1</sup>	RFO <sup>(1</sup>	Furnaces
Residential combustion	-	3.5-5	1.1-1.5	1.1	0.7	$1.4^{(2)}$	5.8
Commercial scale combustion	1.6	0.6			0.7	1.4	

 $^{1}$  RFO = residual fuel oil, DFO = distillate fuel oil.

<sup>2</sup> Also for shale oil.

In other countries (Appendix C),  $CH_4$  IEFs vary from 0.02 to 10 mg/MJ. The Finnish emission factor seems to be at the higher end of the range, and therefore the emission factor could be changed to 4 mg/MJ.

Table 11 presents  $N_2O$  emission factors of the IPCC (1997). In other countries,  $N_2O$  emission factors vary from 0.6 to 12 mg/MJ. These seem rather large when compared with factors presented in literature. The Finnish emission factor could be changed to 1 mg/MJ, which is the same factor as for point sources using burners.

Table 11. Emission factors for  $N_2O$  [mg/MJ] from small-scale combustion of oil from literature.

	Fuel	IPCC, 1997
Residential, oil furnaces	—	0.2
Commercial scale	residual fuel oil / shale oil	0.3
combustion	distillate fuel oil	0.4

#### 2.2.3 Natural gas

Gas combustion, which comprises natural gas and some process gases, covered 14% of methane and 7% of  $N_2O$  emissions from point sources in Finland in 2002. The share has slightly increased since 1990, because of new natural gas plants and enlargement of the supply grid. More than 90% of gaseous fuels are used in large installations (CRF categories 1A1 and 1A2). The most important combustion technologies for natural gas are different gas turbine processes (GTCC, gas turbine with heat recovery etc.). Smaller gas turbines are used to cover peak load, but these gas turbines usually use oil as a fuel. Gaseous fuels are also combusted together with other fuels in multi-fuel fired boilers.

#### 2.2.3.1 CH<sub>4</sub> from point sources (natural gas)

The implied average emission factor for  $CH_4$  from gaseous fuel combustion in point sources (4.5 mg  $CH_4/MJ$ ) was nearly the same in 1990 and 2002. The  $CH_4$  emission from gas combustion is low, due to high combustion temperature and good mixing that enables complete combustion of the gas. The  $CH_4$  emission factors used in the Finnish inventory are given in Table 12. For most installations, emission factor used is 3 mg  $CH_4/MJ$ . Emission factors from different literature sources for natural gas combustion ranged from 0.1 to 240 mg  $CH_4/MJ$  (Table 13). Recent measurements in >100 MW turbines (Appendix B) show that emission factor for gas turbine combustion may be below 1 mg/MJ even with partial load. This factor is very sensitive to disturbances. For example, in the case of an overload running turbine, where natural gas did not mix well with the coming oxygen in supplementary firing, the measured TOC emissions were much higher.

Fuel	Technology/Size	≤5 MW	5–15 MW	15-100 MW	>100 MW
Gas	Burner + grate	_	_	4	3
Gas	Other boilers	3	3	3	3
Gas	Gas turbines	-	3	3	3
Gas	Diesel engine / otto cycle engine	2	2	2	2
Gas 50-80%	Burner	-	3	3	7
Gas 50-80%	LowNo <sub>x</sub> -burner + grate	-	—	15	_
Gas 50-80%	Burner + grate	_	6	_	7
Gas 50-80%	No specification			15	

Table 12. CH<sub>4</sub> emission factors [mg/MJ] for natural gas combustion used in national GHG inventory of Finland.

Table 13 presents  $CH_4$  emission factors for natural gas combustion from literature. In addition, Corinair 1990 presented emission factors 0.3–4 mg/MJ for natural gas

combustion (Corinair, 2004). Tables 16 and 17 present emission factors for VOCs from gas turbines with different NO<sub>x</sub>-control techniques.

	SYKE, 2004	Corina	ir, 2004	IPC	СС, 1997	Measurements
Technology/Size	>15 MW	< 50 MW	> 50 MW	Utility	Industrial	(Appendix B)
Burner/boiler	$1^{(1)}$		0.1-1.4	0.1	1.4 <sup>(2</sup>	
Stationary engine	2	0.02-153		240 <sup>(3</sup>	2.9–17 <sup>(3</sup>	
Gas turbine	3	0.3–23	2.5-6.1	6(4	0.6 <sup>(3</sup>	< 1-26 <sup>(5</sup>

*Table 13.* CH<sub>4</sub> emission factors [mg/MJ] for natural gas combustion from literature.

 $^{1}$  >50% of fuel is natural gas.

 $^{2}$  > 30 MW.

<sup>3</sup> Values for industrial heavy duty gas compressor engines and turbines. For engines factor 17 is for 2-cycle lean burn, 13 for 4-cycle lean burn and 2.9 for 4-cycle rich burn. Factor for utility engines is for dual-fuel engines.

 $^{4}$  > 3 MW.

<sup>5</sup> Factor of 26 mg/MJ represents TOC emissions. It was measured in an occasion where natural gas did not mix well with the coming oxygen in supplementary firing.

IEFs from other countries (Appendix C expect Denmark) vary from 0.07 to 2 mg/MJ for natural gas in public power plants. For Industry, emission factors for natural gas vary from 0.33 to 5 mg/MJ. Denmark reported technology-specific emission factors in the range of 1.5–15, except stationary engines for which EF was 520 mg/MJ.

The Finnish emission factors for boilers and turbines seem to be rather large when compared with the values from literature. Emission factor of 1 mg/MJ could be used for large (> 50 MW) boilers and turbines. Emission factor could be somewhat higher (3 mg/MJ) for small gas turbines, where combustion may be more incomplete or start-ups and shut-downs may play a significant role.

For gas engines the emission factors are much higher than for other combustion technologies. Part of the natural gas goes through the engine unburned which causes high methane emissions. According to Finnish experts (e.g. Riikonen, 2004; Niemi, 2005; Turunen, 2005) emissions from gas engines vary between 100 and 1000 mg/MJ in steady state. There are many different engine types in market and in some types there is less unburned gas in flue gases but this problem is however common in existing power plants. Table 36 presents rough suggestions for emissions factors for different engines based on information from experts. With catalyst, methane emissions are reduced to level of 100 mg/MJ. Therefore this factor could be used in Finland for engines with catalyst.

#### 2.2.3.2 N<sub>2</sub>O from point sources (natural gas)

The implied average  $N_2O$  emission factor which is lowest of all fuels types for gas combustion (1.4 mg  $N_2O/MJ$ ), was nearly the same in 1990 and 2002.  $N_2O$  emission factors for gas combustion used in the Finnish GHG inventory are given in Table 14. Temperatures in gas combustion are usually high and nitrogen of natural gas is molecular like in air, resulting in minor  $N_2O$  emissions. In the inventory, emission factors used are low except diesel engines using gas for which the EF used is rather high (31 mg  $N_2O/MJ$ ). This emission factor is the same as used for oil driven diesel engines, and is likely to overestimate emissions.

Table 14.  $N_2O$  emission factors [mg/MJ] for natural gas combustion used in national GHG inventory of Finland.

Fuel	Technology	<b>Emission factor</b>
Gas 50–100%	Burner + grate	2
Gas 50–100%	Other boilers and gas turbines	1
Gas	Diesel engine	31

For natural gas combustion,  $N_2O$  emission factors from literature varied from 0.1 to 14 mg  $N_2O/MJ$  (Table 15).

Measurements presented in Appendix B show that all three measured  $N_2O$  factors for large (> 100 MW<sub>th</sub>) gas turbines were below the detection limit. Partial load did not have a notable effect on the emission factor.

Technology	SYKE, 2004	IIASA, 2004	Corinair,	2004	Measurements (Appendix B)
			< 50 MW	>50 MW	115–327 MW
Gas turbine	1		0.1–3	0.1-3 <sup>(2</sup>	< 3 (GTCC)
Diesel/gas engine	2	0.1	0.1–3	1–3	

0.1 - 14

Table 15. N<sub>2</sub>O emission factors for natural gas combustion from literature [mg/MJ].

<sup>1</sup>SYKE (2004) suggests that these values should be checked with measurements.

<sup>2</sup> Corinair90 data without technical specification.

Boiler/burner

Grate + burner

0.1<sup>(1</sup>

2

Corinair Emission Inventory Guidebook (2004) presented several measured emission factors for gas turbines with different  $NO_x$ -control technologies. There were no SCR or SNCR among the measured units and the other  $NO_x$ -control technologies did not have a significant effect on the  $N_2O$  emissions (Table 16). Russian natural gas used in Finland does not contain nitrogen, whereas other types of natural gas may contain even 10–15% of nitrogen (Gasum, 1993). However, nitrogen content of combusted natural gas does not have

an effect on  $N_2O$  emission because nitrogen in natural gas is molecular nitrogen (similar as nitrogen in air) that does not react in low temperatures where  $N_2O$  is usually formed.

Table 16.  $N_2O$  and VOC emission factors [mg/MJ] for different  $NO_x$ -control technologies in gas turbines (Corinair, 2004).

Description	NO <sub>x</sub> control system	Number of turbines <sup>(1</sup>	Size [MW]	N <sub>2</sub> O	VOC <sup>(2</sup>
Public power	Low-NO <sub>x</sub> burner	4	150-215	1–7	0.6–2.6
	Steam injection	1	125	< 9	2.6
	Uncontrolled	2	25-125	< 9	1.1-4.2
Industry	Water injection	1	22	2	2.8
	Stem injection	2	4.8–40	2	1.3-6.3
	Uncontrolled	4	6.25–40	≤4	1.6–3.0
Gas extraction/distribution	Uncontrolled	5	6.25–25	2–6	3.7–25

<sup>1</sup>Number of gas turbines in survey.

<sup>2</sup> Expressed as methane.

Corinair (2004) gives also values obtained from literature on the effect on  $NO_x$  control technologies on  $N_2O$  (Table 17). The results are in line with the survey presented above.

Table 17.  $N_2O$  and VOC emission factors for natural gas burning gas turbines equipped with different  $NO_x$ -control technologies (Corinair, 2004).

NO <sub>x</sub> control system	N <sub>2</sub> O emi	ssion factor,	mg/MJ	VOC emission factor, mg/MJ <sup>(1</sup>			J <sup>(1</sup>
	Stewart, 1998	USEPA, electricity	UK NAEI	Stewart, 1998	USEPA, electricity	EMEP/ CORINAIR	UK NAEI
Uncontrolled	2–6			1.1–25	10.3	$\begin{array}{c} 2.5-6.1^{(2)} \\ 2.5-5^{(3)} \end{array}$	
Steam injection	3	1.29		1.3-6.3			
Water injunction	3	1.29		2.8			
Dry. low-NO <sub>x</sub> burner	1–7		3.7	0.6–2.6			6.0 <sup>(2</sup> 5.3 <sup>(3</sup>

<sup>1</sup> Expressed as methane.

 $^{2}$  CH<sub>4</sub>.

<sup>3</sup>NMVOC.

In other countries (Appendix C),  $N_2O$  emission factors for natural gas combustion vary from 0.1 to 3.2 mg/MJ.

In natural gas combustion, temperatures are usually so high that  $N_2O$  emissions should not occur in normal situations. In addition, fuel nitrogen that is in molecular form should not react to  $N_2O$ . Therefore an emission factor of 1 mg/MJ is suggested for all technologies. For other gases, somewhat higher emission factors (2–3 mg/MJ) are suggested due to their varying properties.

#### 2.2.3.3 Small-scale combustion of natural gas

In Finland, small-scale combustion of natural gas has more than doubled since 1990. But its share of energy use in small-scale combustion was only 2% in 2002. Emission factors used are 3 mg/MJ for  $CH_4$  and 1 mg/MJ for  $N_2O$ .  $CH_4$  emission factors for small-scale combustion from literature are presented in Table 18.

Table 18.  $CH_4$  emission factors [mg/MJ] for small-scale natural gas combustion from literature.

	Corinair, 2004	IPCC, 1997
Residential combustion	2.5	—
Residential gas heaters	_	1
Commercial scale boilers	1.2	1.2

Methane IEFs from other countries (Appendix C) show a range from 0.8 to 5 for smallscale natural gas combustion.  $CH_4$  emission factor used in Finland seems high compared with literature sources, and could be changed to 2 mg/MJ.

IPCC (1997) gives an emission factor of 2.3 mg/MJ for  $N_2O$  from natural gas combustion in commercial scale boilers. In other countries (Appendix C), emission factors vary from 0.1 to 2.2 mg/MJ. The emission factor used in Finland seems to fit well with this data.

#### 2.2.4 Biomass

Biomass combustion covers different types of biomass (wood residues from industry, black liquor, biogas etc.). Peat that has traditionally been treated as biomass in Finland, but is not included in this category because it is treated similarly as fossil fuels under the reporting to the UNFCCC. Biomass combustion causes nearly half of  $CH_4$  emissions from combustion in point sources in Finland, and one fifth of N<sub>2</sub>O emissions. Its share of emissions has increased since 1990 (for  $CH_4$  from 34% to 49% and for N<sub>2</sub>O from 18% to 20%) mainly due to increased use of biomass. Most of the biomass (82% in 2002) is combusted in large installations, mainly in pulp and paper industry and public electricity and heat production. Even though the majority of the use is in large installations (CRF 1A1 and 1A2), the emissions (92% in 1990 and 87% in 2002) come mainly from small-scale applications (CRF category 1A4), because combustion in smaller sources is more incomplete than in large boilers. Altogether the share of biomass combustion covered 7% of total methane emissions from Finland in 2002 (3% of N<sub>2</sub>O).

#### 2.2.4.1 CH<sub>4</sub> from point sources (biomass)

Biomass is combusted in Finland using a variety of technologies: mainly recovery boilers and FBC but also grates, burners and gasification. The largest emission factors are estimated for small (< 1 MW) boilers (200 mg CH<sub>4</sub>/MJ). The average implied CH<sub>4</sub> emission factor for biomass is largest of all fuels (9.7 mg CH<sub>4</sub>/MJ in 2002 and 8.1 mg CH<sub>4</sub>/MJ in 1990). Fluidised bed combustion of wood has increased significantly, whereas burning in grates has decreased. The emission factors for point sources used in the 2002 inventory (excluding the CRF category 1A4) are given in Table 19.

Fuel	Technology/Size [MW]	<u>&lt;</u> 1	1–5	5-50	> 50
Wood/bark	FBC, gasification	_	30	30	30
Wood/bark	Other boilers	200	50	50	30
Wood/bark 50-80%	Gasification + air staging	_	_	4	_
Wood/bark 50-80%	FBC	-	35	35	4
Wood/bark 50-80%	Grate	_	35	35	20
Wood/bark 50-80%	Burner + grate	_	50	25	20
Wood/bark 50-80%	Burner	-	_	_	4
Wood/bark 50-80%	Other boilers	_	50	50	_
Black liquor	Recovery boiler	-	-	1	1

*Table 19. CH*<sub>4</sub> *emission factors [mg/MJ] used in national GHG inventory of Finland.* 

Emission factors from literature and measurement reports are presented in Table 20. The IPCC (1997) gives an emission factor of 15 mg/MJ for wood combustion in stoker boilers.

Table 20. Emission factors for biomass combustion from literature [mg CH<sub>4</sub>/MJ].

Technology	Main fuel	<b>SYKE</b> , 2004	1	Corinair, 2004	Measurements
		15-50 MW	> 50 MW		(Appendix B)
> 80% of main f	uel				
FBC	Wood/bark	50	30	15–18 (1	$\leq 1 - 72^{(3)}$
PF/grate	Wood/bark	50	30	15–18 (1	4
Gas turbine	Biogas			2.5	
Not specified (< 50 MW)	Biogas			0.4–10	
Not specified (> 50 MW)	Biogas			0.5–2.5 <sup>(2</sup>	
Recovery boiler	Black liquor			1–18 (2	$\leq 1^{-(4)}$
50-80% of main	fuel				
FBC	Wood/bark	35	4		
Grate/others	Wood/bark	35	20		2
BFB	50% bark, 50% peat				< 1

 $^{1}$  For > 50 MW plants, 15 for industrial and commercial, 18 for utility boilers.

<sup>2</sup> Range as in Corinair (1990) according to Corinair (2004).

Emission factor of 72 mg/MJ was measured in an occasion where air ratio was too low. This 3 emission factor represents emissions of TOC. In other measurements, emissions were  $\leq 1 \text{ mg/MJ}$ .

<sup>&</sup>lt;sup>4</sup> Measurements from recovery boilers with capacity > 2600 tds/d.

In Corinair 1990, emission factors given for point sources combusting wood varied from 1 to 40 mg CH<sub>4</sub>/MJ. Recent measurements (Appendix B) were done for large boilers and the measured CH<sub>4</sub> emissions were very low or even below the detection limit, with one exception. For bark combustion in BFB, high TOC emission (72 mg/MJ) was measured because during the measurements the air ratio was too low. Similar effects have been noted also in some other measurements according to measurement experts of VTT. These results show the sensitiveness of methane emissions to disturbances.

In other countries (Appendix C), emission factors given for biomass combustion vary from 2 to 32 mg/MJ, but Denmark reports emission factors in a more dissaggregated level. Denmark uses an EFs of > 300 mg/MJ for biogas combustion in stationary engines.

The emission factors used in Finland seem rather high for large boilers. In FBC, residence time is usually long enough to prevent CH<sub>4</sub> emissions with good mixing and air ratio. The residence time is longer and technology more advanced for larger furnaces, and therefore an emission factor of 5 mg/MJ is suggested for > 5 MW FBC boilers. For < 5 MW FBC boilers, an emission factor of 10 mg/MJ is suggested. For other furnaces, emission factors can be assumed low in relatively new large scale boilers due to proper air ratio, long residence time, and/or high temperature. However, disturbances and boilers with older technology must also be taken into account in emission factors are suggested for smaller furnaces, i.e. 30 mg/MJ for 5–50 MW and 50 mg/MJ for < 5 MW boilers. There is, however, very little data available from small boilers. CH<sub>4</sub> emission factors for biogas combustion are suggested to be the same as for combustion of other gases than natural gas. In recovery boilers, temperature is very high, and therefore CH<sub>4</sub> emissions are minor (EF of 1 mg/MJ is suggested).

#### 2.2.4.2 N<sub>2</sub>O from point sources (biomass)

The average implied  $N_2O$  emission factor for biomass combustion is rather low, 2.3 mg  $N_2O/MJ$  in 2002. The  $N_2O$  emission factors used in the Finnish inventory are given in Table 21. Largest emission factors are used for circulating fluidised beds (10 mg  $N_2O/MJ$ ).

Table 21.  $N_2O$  emission factors [mg/MJ] for biomass combustion used in the national GHG inventory of Finland.

Fuel	Technology	<b>Emission factor</b>
Wood/bark 50-100%	CFB	10
Wood/bark 50-100%	BFB, other boilers	2
Black liquor	Recovery boiler	1

In general, N<sub>2</sub>O emission is often higher for bark than for wood, but in literature they are usually treated together. Also branches, needles and other parts of tree have different amounts of nitrogen in different forms. Table 22 shows nitrogen content of different parts of tree, explaining why emission factors for pure stem wood and bark should be different.

	Pine	Fir	Birch
Stem wood	0.05	0.05	0.05
Bark	0.3	0.5	0.4
Stem wood with bark	0.1	0.1	0.1
Branches with bark <sup>(1</sup>	0.4	0.5	0.7
Needles	1.1	1.0	_
Spire	0.6	0.8	0.8
Roots ( $d > 0.5$ cm)	0.1	0.1	0.4

Table 22. Typical nitrogen content in different parts of tree [weight-%] (Johansson, 1991).

<sup>1</sup> Without leaves or needles.

Emission factors for biomass combustion found in literature are presented in Table 23. IPCC did not give  $N_2O$  factors for wood combustion.

Table 23.  $N_2O$  emission factors for biomass combustion [mg  $N_2O/MJ$ ] from literature.

Technology and/or size	Fuel	SYKE, 2004	Measurements (Appendix B)	Corinair, 2004
CFB	Wood/bark	10	0–20 (1	
BFB	Wood/bark	2	< 1–2	4.3 <sup>(2</sup>
PF/grate/others	Wood/bark	2		
Grate (< 1 MW)	Wood residues <sup>(3</sup>		< 2	
Recovery boiler	Black liquor		< 1	1–21 (4
< 50 MW	Biogas			1.5-3.7
> 50 MW	Biogas			1.4-2.5

<sup>1</sup> Measurements in Ahlstrom pyroflow CFB boilers gave a typical emission factor of 5–15 mg/MJ in wood combustion (Hiltunen et al., 1991).

 $^2$  For wood combustion in 50–300 MW<sub>th</sub> boilers. For plants < 50 MW emission factors from 1.6 to 20 mg/MJ were presented.

<sup>3</sup> Sawdust, cutter chips, chipboard sawdust.

<sup>4</sup> Expressed as Corinair90 data.

In other countries (Appendix C),  $N_2O$  emission factors for biomass combustion varied from 0.8 to 5 mg/MJ.

According to Finnish combustion and measurement experts the emissions from BFB are negligible for wood combustion, and also emission from CFB are small due to the low nitrogen content of wood. Therefore, for pure wood combustion, emission factor of 1 mg/MJ is recommended. In co-combustion, even small amounts of peat or bark may increase emissions in FBC, and therefore emission factors of 2–4 mg/MJ are suggested

for FBC burning > 80% of wood. For bark combustion, emission factors suggested are 1 for other technologies and 3–10 mg/MJ for FBC depending on technology and significance of partial load. If shares of wood and bark cannot be estimated, emission factors given for bark should be used to be conservative. For biogas, emission factors presented for other gases than natural gas can be used. In recovery boilers, temperature is very high, and therefore N<sub>2</sub>O emissions are minor (EF 1 mg/MJ is suggested).

#### 2.2.4.3 Small-scale combustion of biomass

In Finland, CH<sub>4</sub> emission from small-scale combustion of biomass is a key source in terms of total GHG inventory. Energy input from biomass is slightly smaller than from oil (45% in 2002), but this emission source dominates emissions from small-scale combustion. Emission factors used are 300 mg CH<sub>4</sub>/MJ in the whole time series, and 4 mg N<sub>2</sub>O/MJ in 2002 and 2 mg N<sub>2</sub>O/MJ in 1990. Table 24 presents emission factors from literature.

*Table 24. Emission factors from literature for CH*<sub>4</sub> *from small-scale combustion of biomass.* 

	Corinair, 2004	IPCC, 1997
Residential combustion	74–320	
Wood pits		200
Conventional stoves		210
Catalytic stoves		380

In other countries (Appendix C)  $CH_4$  IEFs vary from 30 to 300 mg/MJ. In 1997, Austria took an emission limit of 80 mg  $C_xH_y/MJ$  at use for wood stoves (Boström et al., 1997). This may indicate that emissions of  $CH_4$  are supposed to be smaller than that. However, according to Finnish experts, burning of wood in households differs from country to country, and therefore emission factors used in other countries may not be applicable for Finland.

 $N_2O$  emission factors in different countries (Appendix C) varied from 1.4 to 7 mg/MJ. Pfeiffer et al. (2000) present  $N_2O$  emission factors of 1.5 mg/MJ for households and 0.99 mg/MJ for other small consumers (see Appendix C).

Methane emissions from households are typically larger than from other small-scale combustion. Currently used emission factor (300 mg/MJ) could still be used for households, but for other combustion an emission factor of 200 mg/MJ is suggested. For N<sub>2</sub>O there is very little data available, but however emission factor could be the same for the whole time series. Emission factor of 3 mg N<sub>2</sub>O/MJ is recommended, because nitrogen content of wood is low.

#### 2.2.5 Peat

IPCC categorises peat under solid fuels in the same category as coal and coal products. In Finland, peat use is reported under "Other fuels", in which peat is the major contributor to the energy production and emissions. Other fuels covered 14% of  $CH_4$  emissions from point sources in 1990, and 9% in 2002 (30% and 27% of N<sub>2</sub>O). The significance of peat combustion to the total N<sub>2</sub>O emissions in Finland is around 3%, but the significance of peat combustion to total  $CH_4$  emissions from Finland is only 0.2%. Use of peat has increased, and the decrease in emissions is due to change in technology and decrease in emission factors.

Majority of peat is combusted in large installations (CRF 1A1 and 1A2), 99% in 2002 and 98% in 1990. In Finland, peat is combusted mainly using FBC. The use of FBC, especially BFB, has increased significantly. Peat is often combusted together with, e.g., coal and biomass. To some extent, peat is also combusted using pulverised firing, and grates are used in smaller boilers. There is also minor use of peat with gasification technology. Some properties of peat have changed during the last decades. This may have led to changes also in emissions. For example average moisture of supplied peat has decreased 2–3 mass-% from 1990 to 2003 (Vapo, 2005).

#### 2.2.5.1 CH<sub>4</sub> from point sources (peat)

Implied emission factor for all  $CH_4$  emissions from "Other fuels" has decreased from 8.1 mg/MJ to 4.5 mg/MJ between 1990 and 2002.  $CH_4$  emission factors for peat combustion used in the Finnish inventory are given in Table 25.

*Table 25.* CH<sub>4</sub> emission factors for peat combustion used in national GHG inventory of Finland [mg/MJ].

Fuel	Technology/Size [MW]	<u>&lt; 1</u>	1–5	5–15	15-50	50-150	> 150
Peat	Grate, burner + grate	50	5	2		7	
Peat	FBC, gasification, burners, others	_	5	5	5	7	2
Peat 50-80%	Burner	_	5	_	_	_	2
Peat 50-80%	Burner + grate	_	_	_	9	_	_
Peat 50-80%	Grate	_	5	2	15	15	-
Peat 50-80%	BFB	_	_	5	-	2	2
Peat 50-80%	BFB + burner	_	_	_	_	15	_
Peat 50-80%	CFB, other boilers	_	5	_	5	5	5

Emission factors given for peat combustion in other sources are presented in Table 26. The IPCC guidelines (1997) did not provide  $CH_4$  emission factors for this source.

Technology/	SYKE,	2004	Corinair, 2004		Measurements
Size [MW]	15-50	> 50	< 50	50-300	(Appendix B)
Pulverized firing		2			
CFB	5	1			≤ 1 (> 100 MW)
BFB	3	2	5–400	1-39 (1	2-3 <sup>(2</sup> (78-260 MW)
Mixed combustion (50–80% peat)	15	2			< 1 (78 MW)
Grate	2	2			

Table 26. Emission factors for methane from peat combustion [mg/MJ].

<sup>1</sup> Presented in Corinair (2004) as Corinair90 data.

<sup>2</sup> Emission factor represents emissions of TOC.

In Sweden (Appendix C), IEF for peat combustion is 20 mg/MJ.

Methane emission factors used for large grates as well as FBC, gasification and burners are larger than for corresponding smaller combustion technologies in the Finnish GHG inventory. This is clearly a mistake and should be corrected. The magnitude of the emission factors used corresponds largely to emission factors derived from measurements and literature. However, for large boilers (> 50 MW) annual average emission factors are too high compared to the other sources. Emission factor of 3 mg CH<sub>4</sub>/MJ is more suitable for this category. For smaller units, emission factors from 3 to 50 mg/MJ are suggested (see Table 36).

#### 2.2.5.2 N<sub>2</sub>O from point sources (peat)

Implied emission factor for all  $N_2O$  emissions from "Other fuels" has decreased from 12 mg/MJ to 8 mg/MJ between 1990 and 2002. The decrease in the implied emission factor is due to the change in the technology as well as increased co-firing with wood. In addition, some inconsistencies in time series occur. Nitrogen content of peat varies between 0.6 and 3.0 percent of dry solids depending on the type of soil where peat has been extracted. Therefore, emission factors for peat combustion may be different in each country and even in different regions of a country. Typically, nitrogen content is higher for peat from northern than southern Finland. Nitrogen content of peat has not changed significantly since 1990. If peat is presently combusted in different regions of Finland than in 1990, then the average nitrogen content of combusted peat may be different (Vapo, 2005).

The  $N_2O$  emission factors used in the Finnish inventory are given in Table 27 and  $N_2O$  emission factors for peat combustion from literature are presented in Table 28.

Table 27.  $N_2O$  emission factors [mg/MJ] for peat combustion used in the national GHG inventory of Finland.

Fuel	Technology	EF
Peat 50-100%	CFB	30
Peat 50-100%	BFB, other boilers	2

Table 28.  $N_2O$  emission factors for peat combustion from literature [mg  $N_2O/MJ$ ].

Technology	SYKE,	Corinair, 2004		Measurements
	2004	< 50 MW	> 50 MW	(Appendix B)
CFB	15			< 2–18 <sup>(1</sup>
BFB	2	2-14	2-75 (2	< 2
Grate/PF/others	2			$22^{(3)}$

 $^1$  Measurements with full load. In Appendix B, factors for partial load are also presented. In Ahlstrom pyroflow CFB boilers, 5–15 mg/MJ are given as typical  $\rm N_2O$  emission factors in peat combustion (Hiltunen et al., 1991).

<sup>2</sup> Expressed as Corinair90 data.

 $^{3}$  < 1 MW intermittent grate, in the beginning of 90's.

An emission factor of 26 mg/MJ was measured for peat combustion in a CFB plant with partial load (75%) (Appendix B). This high value shows the sensitiveness of the emission to the combustion conditions (a load of 75% decreases the combustion temperature). See Chapter 2.1.4 for more analysis about partial load. In Ireland and Sweden, IEFs for peat combustion vary from 10 to 12 mg/MJ.

The emission factor used for BFB in the Finnish inventory is in line with measured data and values presented in literature (EF of 3 mg/MJ is recommended). The emission factor used for CFB may be too high. Therefore, an emission factor of 20 mg/MJ is recommended for district heating CFB and 15 mg/MJ for other CFB. For other technologies, emission factor of 2 mg/MJ is suggested. The impact of the load and other disturbances as well as that of co-firing peat with other fuels should be studied further.

The only measurement result for small grate shows large  $N_2O$  emissions in the beginning of 1990's. This may reveal that 1) emissions are higher for smaller grates than larger grates and/or 2) emissions were higher in the beginning of 1990's than today due to differences in boiler driving and maintenance. However, no conclusions can be made based on a single measurement result.

#### 2.2.5.3 Small-scale combustion of peat

Use of peat in small-scale combustion is decreasing in Finland. In 2002, the amount of peat (as energy) combusted in small-scale combustion was half of the value in 1990.

 $CH_4$  emission factor used was 200 mg/MJ in 1990 and 50 mg/MJ in 2002.  $N_2O$  emission factor was 4 mg/MJ in 2002 and 2 mg/MJ in 1990.

In Ireland,  $CH_4$  IEF is 50, and in Sweden 30 mg/MJ. N<sub>2</sub>O IEFs are 5 and 10 mg/MJ, respectively. There is no literature or other data to support the change of Finnish emission factors. However, the factors for small-scale combustion should be the same for the whole time series, if there is no good reason for the change.

#### 2.2.6 Waste

Waste combustion is reported in Finland under Other fuels except biomass wastes like industrial wood residues that are reported under Biomass. Wastes reported under Other fuels cover, e.g., industrial waste, plastic waste, MSW and REF. In Finland, waste is typically combusted with other fuels, e.g. to substitute 5–30% of fossil fuels in a boiler. In large CHP plants, waste is often combusted using fluidised bed combustion, but in small heating plants waste is typically combusted on grates. Waste fuels are also produced by pelletizing and using gasification in fluidised beds. Practically all combusted waste is used in large installations (CRF categories 1A1 and 1A2). The total amount of waste combusted in 2002 was around 5000 TJ (2% of total Other fuels) and in 1990 about 1200 TJ. Methane emissions from waste combustion were 0.038 Gg in 2002 (0.018 Gg in 1990), and the corresponding IEF 8 mg CH<sub>4</sub>/MJ. N<sub>2</sub>O emissions from waste combustion were 0.026 Gg in 2002 (0.004 Gg in 1990).

The wet LHV of typical combusted waste (including MSW, RDF, demolition wood and industrial waste) varies between 11 and 18 MJ/kg. Variation is mainly due to moisture content of the fuel. LHV of wet sludge is smaller in magnitude because water content can be around 80%. Nitrogen contents of these fuels are 0.8–1.4% of dry solids which is smaller than N-content of e.g. peat. Exception is sludge which nitrogen content is high, about 3.3% (Rautanen, 2004).

#### 2.2.6.1 CH<sub>4</sub> from point sources (waste)

Majority of wastes combusted in Finland are combusted in co-firing with other fuels. Therefore the emission factors correspond with emission factors of the main fuel used in the boiler, or can be calculated as described in Section 2.1.3. In the Finnish GHG inventory,  $CH_4$  emission factors for hospital waste incineration are 50 mg/MJ for ovens < 5 MW and 8 mg/MJ for ovens > 5 MW.

 $CH_4$  emission factors for waste combustion given in (Corinair, 2004) varied from 1 to 400 mg  $CH_4/MJ$  depending on waste type and combustion technology (Table 29). The IPCC (1997) did not give  $CH_4$  emission factors for this source.

Table 29. Methane emission fo	actors [mg CH4/MJ] for waste o	combustion (Corin	air, 2004).
Sizo	> 50 MW	< 50 MW	

Size	> 50	< 50 MW	
Fuel/Type	No specification <sup>(1)</sup>	Commercial	No specification
Municipal waste	1	6.5 <sup>(2</sup>	6–32
Industrial waste	10		0.3–38
Wood waste	4–40		30–400
Agricultural waste	32	9 <sup>(2</sup>	10-400
Waste gas	2.5		0.4–2.5

<sup>1</sup> Expressed as Corinair90 data in Corinair (2004) without technical specification. <sup>2</sup> Open burning.

Other countries (Appendix C) presented IEFs from 0.06 to 20 mg/MJ for MSW. Recommended emission factors (Table 36) for waste combustion are 10 mg/MJ for < 5 MW plants, 3 mg/MJ for 5–50MW, and 1 mg/MJ for large (> 50 MW) plants.

### 2.2.6.2 N<sub>2</sub>O from point sources (waste)

Majority of wastes combusted in Finland are combusted in co-firing with other fuels. Therefore, the emission factors correspond with emission factors of the main fuel used in the boiler, or can be calculated as described in Section 2.1.3. For hospital waste incineration, Finland uses emission factor of 2 mg N<sub>2</sub>O/MJ. N<sub>2</sub>O emission factors for waste incineration from (Corinair, 2004) are presented in Table 30.

Table 30. $N_2O$ emission	factors for waste in	ncineration [mg ]	N2 <b>O/MJ</b> ] ((	<i>Corinair</i> , 2004).
	J	L	- 2	

Fuel	< 50 MW	> 50 MW <sup>(1</sup>
Municipal waste	4	4 <sup>(2</sup>
Industrial waste	2-5.9	1.4
Wood waste	4	2-6
Agricultural waste	1.4–4	5
Waste gas	3.7–5	1.1–2.5

<sup>1</sup> Expressed as Corinair90 data in Corinair (2004) without technical specification.

<sup>&</sup>lt;sup>2</sup> For MSW, factors 14–165 g/t waste for FBC and 11–270 g/t waste for grate firing were also given. With typical heating value of dried MSW, 12 MJ/kg (Lohiniva et al., 2001) the calculated emission factors are 1–14 and 1–23 mg/MJ.

Other countries (Appendix C) presented  $N_2O$ -emission factors from 1.2 to 5 mg/MJ. Recommended emission factors (Table 37) for waste combustion are 10–15 mg/MJ for CFB, 5 mg/MJ for BFB and 4 mg/MJ for other technologies.

#### 2.2.7 Mixed fuels

This category includes co-combustion where no specification of main fuel has been provided, because share of each fuel type is < 50%.

#### 2.2.7.1 CH<sub>4</sub> from point sources (mixed fuels)

 $CH_4$  emission factors used in the Finnish inventory for mixed combustion and for combustion with no specification on fuel type are presented in Table 31.

Table 31.  $CH_4$  emission factors [mg/MJ] used in the Finnish GHG inventory for cocombustion of mixed fuels (share of each fuel < 50%).

Technology/Size [MW]	<u>&lt;</u> 1	1–5	5–15	15-50	50-150	> 150
Grate, burner + grate	_	35	_	15	10	-
BFB	_	_	_	15	15	15
Burner	50	35	6	4	6	7
CFB with air staging	_	30	_	_	_	_
No specification	50	50	15	15	15	7

SYKE (2004) presents the following  $CH_4$  emission factors (Table 32) for fuel mixes which contain less than 50% of each fuel component.

Table 32.  $CH_4$  emission factors [mg/MJ] for co-firing (SYKE, 2004). These values are largely derived from the values used in the Finnish GHG inventory.

Technology/Size	15-50 MW	> 50 MW
Burner	3	2
CFB, BFB	15	4
Grate, others	15	10

Guidance for estimating emission factors for co-firing of different fuels is given in Section 2.1.3.

#### 2.2.7.2 N<sub>2</sub>O from point sources (mixed fuels)

The  $N_2O$  emission factors used in the Finnish inventory for mixed combustion are given in Table 33.

Table 33.  $N_2O$  emission factors [mg/MJ] used in the Finnish GHG inventory for mixed combustion.

Technology	EF
CFB	30
BFB, other boilers	2

SYKE (2004) presents the following  $N_2O$  emission factors (Table 34) for fuel mixes which contain less than 50% of each fuel component.

Table 34.  $N_2O$  emission factors [mg/MJ] for mixed combustion in > 15 MW applications (SYKE, 2004). These values are largely derived from the values used in the Finnish GHG inventory.

Technology	EF
CFB	15
BFB	2
Grate, others	2

Guidance for estimating emission factors for co-firing of different fuels is given in Section 2.1.3. If there is lack of knowledge on the fuels combusted, emission factors given for peat may be a good approximation. Most common fuel mixes in co-combustion are wood and peat, and  $N_2O$  emission factors given for peat are between emission factors for wood and coal.

#### 2.2.8 Others (e.g. kilns, ovens and dryers)

This category includes combustion in, e.g., lime kiln, asphalt mixing plant, brick kiln, drying oven and drying plant. In the case of these emission sources, emission factors used in the inventory are determined by the process type rather than by fuel. The temperatures in kilns and ovens are usually high and  $CH_4$  and  $N_2O$  emissions have therefore been estimated to be low.

#### 2.2.8.1 CH<sub>4</sub> from point sources (others)

The  $CH_4$  emission factors used in the Finnish inventory for "other" sources are given in Table 35. IPCC gives emission factors 1.0 mg/MJ for coal and oil and 1.1 mg/MJ for natural gas burning in kilns and dryers.

Table 35.  $CH_4$  emission factors [mg/MJ] for "other combustion" used in the national GHG inventory of Finland.

Туре	Emission factor
Coking plant, blast furnace	0
Sintering plant	4
Rolling mill, melting stoves	0
Cupola furnace	8–10
Other combustion process	8–10
Other combustion <sup>(1</sup>	8

<sup>1</sup> E.g. lime kiln, asphalt mixing plant, brick kiln, drying oven and drying plant.

Temperatures e.g. in lime and brick kilns and melting stoves are high enough to enable complete oxidation of methane. Therefore emission factor of 1 mg/MJ could be used for these processes. IPCC emission factors (1 and 1.1 mg/MJ) support this suggestion.

#### 2.2.8.2 N<sub>2</sub>O from point sources (others)

In the Finnish GHG inventory, N<sub>2</sub>O emission factor of 2 mg/MJ is used for lime kilns, coking plants, blast furnaces, drying ovens, drying plants, sintering plants, rolling mills, melting stoves, asphalt mixing plants, brick kilns and cupola furnaces. In lime and brick kilns temperature in the furnace is high, and therefore N<sub>2</sub>O emissions are small. An emission factor of 1 mg/MJ could be used for lime and brick kilns, and currently used emission factor (2 mg/MJ) could be used for other processes.

## 2.3 Recommendations for $CH_4$ and $N_2O$ emission factors

#### 2.3.1 Emission factors

In this Section, recommendations for emission factors for the Finnish GHG inventory are given based on data presented in the previous Sections. Recommended emission factors for methane from point sources are presented in Table 36. Emission factors given for engines using natural gas are rough estimates because emissions vary largely between different engine types. Therefore, one possibility would be to use the IPCC

(1997) emission factor 240 mg/MJ given for dual-fuel engines instead of the values given in the table below.

		Size [MW]				
Fuel	Technology	< 5	5–50	> 50		
Coal > 80%	PF, FBC	4	1 (0.6–2.4)	1 (0.6–2.4)		
Coal > 80%	Other	4	4 (0.7–4)	1 (0.7–4)		
Oil > 80%	Furnaces, FBC	4	4	1 (0.2–3)		
Oil > 80%	Gas turbine	3	3 (1-8)	1 (1-8)		
Oil > 80%	Engines	4	4	2		
Gas > 80%	Furnaces, FBC, GTCC	1 (0.1–1.4)	1 (0.1–1.4)	1 (0.1–1.4)		
Gas > 80%	Gas turbines	3 (0.3–6)	1 (0.3–6)	1 (0.3–6)		
Gas > 80%	Otto engines (without catalyst)	850 (100–900)	800 (100–900)	800 (100–900)		
Gas > 80%	Dual-fuel engines (without catalyst)	650 (200–900)	600 (200–900)	600 (200–900)		
Gas > 80%	Engines with catalyst	100	100	100		
Wood & bark > 80%	FBC	10	5 (0.5–50)	5 (0.5-50)		
Wood & bark > 80%	Other furnaces	50	30 (15-50)	5		
Black liquor	Recovery boiler	_	1	1		
Peat > 80%	FBC	10	3 (1–5)	3 (1-5)		
Peat > 80%	Other furnaces	50	10	3		
Waste > 80%	No specification	10	3	1		

*Table 36. Recommended emission factors [mg/MJ] for methane from point sources. Typical ranges of emission factors are in brackets.* 

For N<sub>2</sub>O, emission factors for > 5 MW plants are presented in Table 37. There is not enough information to provide N<sub>2</sub>O emission factors for smaller boilers, but some studies show that they should be larger due to shorter residence time in furnace and less advanced technology. Also moist fuels may have greater effect on temperatures in small furnaces and therefore N<sub>2</sub>O emissions may be higher.

	Technology							
Fuel	BFB	CFB, district heat	CFB, Other	Burner	Gas turbines	Engines	Others	
Coal > 80%	20 (14–96)	60 (14–96)	30 (14–96)	1 (0.5–1.7)	_	_	3 (0.8–4)	
Oil > 80%	20	30	25	1	4 (0.6–16)	4 (1–7)	4	
Natural gas > 80%	1	1	1	1(0.1-2.4)	1 (0.1–3)	1 (0.1–3)	1 (0.1–3)	
Other gases > 80%	3	3	3	2	2	2	2	
Wood ~100%	1	1	1	1	-	-	1	
Wood > 80%	2	4	3	1	-	-	1	
Bark > 80%	3 (< 1–3)	10 (5-20)	5 (5-20)	1	_	_	1	
Peat > 80%	3	20 (< 2–20)	15 (< 2–15)	2	_	_	2	
Waste > 80%	5	15	10	4	_	_	4	
Black liquor		Recovery boiler, 1						

Table 37. Recommended emission factors [mg/MJ] for  $N_2O$  from point sources (> 5 MW). Typical ranges of emission factors are in brackets.

Table 38 gives recommendations for industrial sources (e.g. lime kilns, blast furnaces), where emission factor is given by process type rather than by fuel used. These emission factors could be used for these processes even if the fuel is known.

Table 38. Recommended emission factors for industrial combustion sources, where emission factor is specified by process type rather than by fuel used.

Туре	CH <sub>4</sub> [mg/MJ]	N <sub>2</sub> O [mg/MJ]
Lime kiln, brick kiln	1	1
Coking plant, blast furnace, sintering plant	1	2
Drying oven, drying plant, rolling mill, melting stove, cupola furnace	1	2

Emission factors for other co-combustion than given in the tables below can be developed based on fuel-specific emission factors and guidance given in Chapter 2.1.3. For gasification and combustion of product gases emission factors given for gas (CH<sub>4</sub>) and other gases (N<sub>2</sub>O) should be used. These are the most suitable factors also for e.g. blast furnace gas and coke oven gas combustion. For wood wastes emission factor should be estimated case by case depending on the composition of the fuel (e.g. share of bark, wood and possible chemicals like glue). For LPG the emission factors of oil could be used. If there is a catalysator (e.g. SCR) in the plant, both emission factors (CH<sub>4</sub> and N<sub>2</sub>O) may be significantly lower. SNCR may increase N<sub>2</sub>O emissions and therefore small increase in emission factor could be assumed if there is a SNCR in the process.

Recommended emission factors for small-scale combustion are given in Table 39.

Table 39. Recommended emission factors for small-scale combustion [mg/MJ] accompanied with possible range of emission factor.

Fuel	Туре	CH <sub>4</sub> EF (possible range)	N <sub>2</sub> O EF (possible range)
Coal	Residential	250 (150-450)	6 (1–20)
Coal	Other heating of buildings	10	6 (1–20)
Oil	All	4 (0.02–6)	1 (0.6–2)
Gas	All	2 (0.12–2.5)	1 (0.1–2.3)
Biomass	Residential	300 (74–380)	3 (1–7)
Biomass	Other heating of buildings	200	3 (1–7)
Peat	All	50	4

Based on interviews of experts, emission factors may have changed radically since 1990. For example, better adjusting of air ratio has possibly decreased the average  $CH_4$  emission factor by an order of magnitude, and changes in plant design and maintenance may have led to decreasing N<sub>2</sub>O. But, there is not any quantitative information available to back up these arguments. Therefore, we recommended using emission factors given in the tables above for the entire time series.

#### 2.3.2 Uncertainties

Calculation of emissions from stationary combustion is carried out at a very detailed level, i.e. plant or boiler specific level for point sources. If the uncertainty analysis is to be carried out at the same level of detail, data of uncertainty in each emission factor should be estimated at plant-specific level. This is because even though emissions are estimated at a plant-specific level, emission factors are given for larger groups, as presented in Tables 36–39. Due to variation between plants in each category, suitability of emission factors used for each individual plant can be different. On the other hand, possible bias in emission factors is likely to correlate across plants. In addition, emission factors in different categories correlate to some extent, because they are partly based on the same data: many emission factors are taken from the same measurement reports; same experts have estimated possible emission factors from a theoretical basis for a number of technologies; and scarce measurement data has been generalised to cover many sub-categories or size-classes. Therefore, for the estimation of uncertainty, correlations between emissions factors applied to different plants should also be assessed, which would be a challenging task.

In the recent inventories (2001–2003), uncertainty estimates presented in Table 40 have been used for  $CH_4$  and  $N_2O$  emission factors for stationary combustion. Estimates are done rather roughly at the third CRF level. In addition, perfect correlation has been

assumed for implied emission factors for the same fuel type across categories 1A1 and 1A2. In this disaggregation level, other correlations could mainly be avoided.

<b>CRF</b> Category	Fuel type	GHG	Uncertainty <sup>2</sup>	Distribution
$All^1$	Liquid fuels	$\mathrm{CH}_4$	-75 to +10%	Beta
All <sup>1</sup>	Liquid fuels	N <sub>2</sub> O	-75 to +10%	Beta
$All^1$	Solid fuels	$CH_4$	-75 to +10%	Beta
$All^1$	Solid fuels	N <sub>2</sub> O	±50	Normal
$All^1$	Gaseous fuels	$CH_4$	-75 to +10%	Beta
$All^1$	Gaseous fuels	N <sub>2</sub> O	±50	Normal
1A1, 1A2	Biomass	$CH_4$	±50	Normal
1A1, 1A2	Biomass	N <sub>2</sub> O	-70 to +150%	Lognormal
1A4, 1A5	Biomass	$CH_4$	-70 to +150%	Lognormal
1A4, 1A5	Biomass	N <sub>2</sub> O	-70 to +150%	Lognormal
$All^1$	Other fuels	$\mathrm{CH}_4$	±50	Normal
$All^1$	Other fuels	$N_2O$	-70 to +150%	Lognormal

Table 40. Uncertainty estimates used in the Finnish GHG inventory for stationary combustion for 2001–2003 inventories.

<sup>1</sup>CRF 1A1, 1A2, 1A4 and 1A5.

<sup>2</sup> Upper and lower bounds of 95% confidence interval relative to the mean value.

Due to the reasons explained above, we recommend the use of the same rough aggregation level for uncertainty estimate also when the emission factors recommended by this report are taken at use, unless more detailed analysis of cross-correlations is available. But, some changes to the previous uncertainty estimates are recommended. Especially, negatively skewed uncertainties that were used to describe the potentially too high emission factors, can be avoided when the emission factors recommended in this report are taken at use. The uncertainties to be used with the emission factors of this report are presented in Table 41. The uncertainty estimates presented in the table represent uncertainty of the total emissions in a given category for a specific fuel type. Relative uncertainties in individual emission factors and in emissions of individual plants may be larger. Uncertainty in each emission factor can be estimated based on possible ranges of emission factors given in Tables 36–39.

Table 41. Recommended uncertainties to be applied for each CRF category at third CRF level (1A1, 1A2, 1A4, 1A5) separately when the emission factors recommended in this report are taken at use.

Fuel type	GHG	Uncertainty <sup>1</sup>	Distribution
Liquid fuels	CH <sub>4</sub>	±50	Normal
Gaseous fuels	CH <sub>4</sub>	±50	Normal
Solid fuels	CH <sub>4</sub>	-70 to +150%	Lognormal
Biomass	CH <sub>4</sub>	-70 to +150%	Lognormal
Other fuels	CH <sub>4</sub>	-70 to +150%	Lognormal
Liquid fuels	N <sub>2</sub> O	-70 to +150%	Lognormal
Gaseous fuels	N <sub>2</sub> O	±50	Normal
Solid fuels	N <sub>2</sub> O	-70 to +150%	Lognormal
Biomass	N <sub>2</sub> O	-70 to +150%	Lognormal
Other fuels	N <sub>2</sub> O	-70 to +150%	Lognormal

<sup>1</sup> Upper and lower bounds of 95% confidence interval relative to the mean value.

# 3. Recommendations for further work in CH<sub>4</sub> and N<sub>2</sub>O emission factors for stationary combustion

Emission factors presented in Chapter 2 were based on literature survey and expert knowledge. Some emission factors still need further development. Especially partial load and co-firing seem to be important issues, of which there is not enough data available for the development of emission factors. There are needs for both measurements and data collection in these areas.

## 3.1 Sensitivity analysis of small-scale combustion

According to emission factors used in 2002,  $CH_4$  emissions from small-scale combustion were 14 Gg and N<sub>2</sub>O emissions 0.3 Gg. The key category analysis for the small-scale combustion identified 5 most important emission categories presented in Table 42. In the total Finnish GHG inventory,  $CH_4$  from biomass combustion in small-scale applications was identified as a key in 2002.

Fuel	Туре	Gas	Emissions in 2002 (Gg CO <sub>2</sub> -eq)	Level assessment	Cumulative level assessment
Wood	Residential	$CH_4$	265	0.70	0.70
Wood	Residential	N <sub>2</sub> O	52	0.14	0.83
LFO	Residential	N <sub>2</sub> O	22	0.06	0.89
Wood	Other	CH <sub>4</sub>	16	0.04	0.93
LFO	Residential	CH <sub>4</sub>	7	0.02	0.95

Table 42. Key categories under small-scale combustion in 2002.

Methane emission factor for wood combustion in small-scale applications is very uncertain. If it was changed to 100 mg/MJ (inside values given in literature), emissions from small-scale applications would decrease around 50% in both 1990 and 2002. In terms of non-CO<sub>2</sub> emissions from stationary combustion this would mean a decrease of 13% and in total inventory (including all sectors and gases) a decrease of 0.2%.

## 3.2 Recommendations for measurements

 $CH_4$  emissions are usually very small in efficient combustion in large installations, but are larger in small-scale combustion (CRF 1A4). The emission factors used for this category are the IPCC default values which are very coarse. The technological

development in the small-scale combustion equipment may not be reflected in these emission factors. Therefore, measurements in this size class would improve the accuracy of the inventory.

During 2005, a measurement project will be carried out at VTT Processes, where emissions from small-scale combustion will be measured, and the results will be publicly available. This project will give more information on  $CH_4$  and  $N_2O$  emissions from small-scale combustion.

In small-scale combustion and furnaces smaller than 10 MW, effect of fuel moisture on  $CH_4$  and  $N_2O$  emissions is unclear. Moist fuels decrease combustion temperature and therefore, it is possible that emissions increase. Therefore, information on the effect of moisture of fuels on emissions should be studied further.

 $N_2O$  emissions from combustion of peat are a key source in the inventory. The emission factors used contain still much uncertainty although the measurements done by Korhonen et al. (2001) have improved the knowledge considerably. For example effect of load changes on  $N_2O$  emissions in fluidised bed combustion needs to be studied further.

In addition, co-firing of peat and wood, and other fuels has increased. The emission factors for the fuel mixes are largely unknown. Measurements would be needed to improve the knowledge and to establish estimation methods (formulas) for determining emission factors for different types of fuel mixes. The measurements should be combined with a study of the influence of load on the emission factors. Especially measurements on CHP plants would be useful. The measurements should cover typical operating conditions throughout the year (e.g. start-ups and disturbances). There is contradictory information about the effect of limestone feeding to furnace in the literature. Method is common in FBC and therefore it could be studied further. In addition, measurements before and after SNCR and SCR of operating plant would give important information about the effect of these devices on emissions.

## 3.3 Recommendations for further data collection

Partial load seems to be an important factor for  $N_2O$  emissions from FBC. In addition to measurements of emissions in specific situations, data on the occurrence of partial load in the scale of Finland should be collected to be able to develop more representative emission factors. Some data about occurrence of partial load may also be available for the year 1990. Co-firing of different fuels is another important issue. There is a need for both measurement data of emissions, and data collection of the typical fuel mixes used in Finnish power plants.

In addition, significance of the changes in  $NO_x$ -reducing technologies, automation and combustion control equipment should be further investigated. This could be done for example by measurements in plants with older automation, air ratio control and technology. Data collection about use of air ratio control by flue gas measurements in plants since 1990 would also improve knowledge of the emissions. Furthermore, CO-levels of measurements done during last two decades could be used for the estimation of changes in CH<sub>4</sub> emission levels.

# 4. Industrial processes – $CH_4$ and $N_2O$

In Finland, Industrial processes emit all greenhouse gases of the Kyoto Protocol, of which  $CO_2$  and f-gases are beyond the scope of this work. In 2002, the only industrial key source (except f-gases) was N<sub>2</sub>O from nitric acid production. Table 43 summarises the currently used emission factors.

Table 43. Emission factors currently used for  $CH_4$  and  $N_2O$  emissions from industrial processes.

Emission source	<b>Emission factor</b>	Unit	Source
Nitric acid production	9.2–9.7	kg N <sub>2</sub> O/t nitric acid	Measurement data
Ethylene production	1	g CH <sub>4</sub> /kg ethylene	IPCC, 1997
Coke Production	0.5	kg /t coke	IPCC, 1997

## 4.1 Nitric acid production (CRF 2B2)

Nitric Acid production releases N<sub>2</sub>O as by-product of catalytic oxidation of ammonia. Nitric acid production includes the following steps:

- 1. catalytic oxidation of ammonia (NH<sub>3</sub>) with air into nitric oxide (NO)
- 2. oxidation of nitric oxide into nitrogen dioxide (NO<sub>2</sub>)
- 3. absorption of nitrogen dioxide in water to produce nitric acid.

Formation of  $N_2O$  in this process depends only on the first step. In catalytic oxidation of ammonia, possible N-products include (in increasing order of oxidation)  $N_2$ ,  $N_2O$  and NO. Degree of oxidation of ammonia depends mainly on the temperature but also on other oxidation conditions, catalyst composition and age and burner design. Once nitrous oxide is formed, it usually passes unreacted through the plant (Pérez-Ramírez et al., 2003).

In nitric acid plants,  $N_2O$  concentration of off-gas is usually 500–2000 ppm. Most nitric acid plants have to regulate their  $NO_x$  emissions, and use therefore SCR or NSCR. NSCR can reduce  $N_2O$  up to 90%, but requires additional fuel and thus increases energy costs (IEA GHG R&D, 2000).

 $N_2O$  emissions from nitric acid production plants depend strongly on plant design, operation conditions, maintenance as well as  $NO_x$  and  $N_2O$  reduction technologies. Table 44 summarises emission factors presented by different sources.

Country/		<b>DD</b> (1	<i></i>
region	Technology	EF <sup>(1</sup>	Source
Austria		4.97-5.30 <sup>(2)</sup>	Radunsky et al., 2003 <sup>(3)</sup>
Canada	European design, without NSCR	8.5 <sup>(4</sup>	IPCC, 2000
Canada	With NSCR	< 2	IPCC, 2000
Europe	Plants with SCR	1.14	IIASA, 2004
Europe		5.7	IIASA, 2004
European design	Dual pressure, double absorption	8–10	IPCC, 2000
Finland		9.2–9.7	Measurement data
Japan		2.2-5.7	IPCC, 2000
Netherlands		9	Oliver et al., 2003 <sup>(3)</sup>
Norway	Medium pressure	6–7.5	IPCC, 2000
Norway	Atmospheric pressure	4–5	IPCC, 2000
Norway	Process-integrated N <sub>2</sub> O destruction	< 2	IPCC, 2000
Sweden		4.8–7 <sup>(2</sup>	Feldhusen et al., 2004 <sup>(3)</sup>
UK	Medium pressure plant	7	Baggott et al., 2004
USA	Without NSCR	9.5 <sup>(4</sup>	IPCC, 2000
USA	With NSCR	1.12-2.5	IPCC, 2000
USA		2–9	IPCC, 1997
World		25	Oliver et al., 1999
World	No abatement	9.5–10	IEA GHG R&D, 2000
_	Older plants (before 1975) without NSCR	10–19	IPCC, 2000

*Table 44. Emissions factors for N<sub>2</sub>O from nitric acid production from different sources.* 

 $^{1}$  Kg N<sub>2</sub>O/t nitric acid.

<sup>2</sup> Range represents variation in IEF between 1990 and 2002.

<sup>3</sup> Company/plant specific data.

<sup>4</sup> Reported uncertainty  $\pm 10\%$ .

In Finland, nitric acid is produced in three medium-pressure plants (1 in Siilinjärvi and 2 in Uusikaupunki). Nitric acid production plant in Oulu was shut down in 1992. Current plants have Selective Catalytic  $NO_x$  Reduction (SCR). Measurements to determine the level of emissions have been done at all plants during 1999, 2000 and 2001. The number of measurements is low, and the uncertainties in emission factors based on measurements are still estimated to be high. There are plans to start continuous on-line measurements at the plants. These results could be used directly in the inventory in the future.

At present, the inventory is based on the plant-specific emission factors based on a set of measurements done in 1999 (average value of the EF in 2002 was 9.4 kg N<sub>2</sub>O/t nitric acid). Incorporation of the data on all measurement would improve the emission estimates. The use of future data from online measurements would improve the accuracy of the estimates further.

#### 4.2 Ethylene production (CRF 2B5)

For CH<sub>4</sub> emissions from ethylene production, Finland uses IPCC (1997) default emission factor 1 g CH<sub>4</sub>/kg ethylene. Oliver et al. (1999) provide an emission factor of 0.25 g CH<sub>4</sub>/kg ethylene. Corinair (2004) gives EFs from 0.6 to 10 kg VOC/t ethylene. Based on few estimates on the share of methane of VOC from ethylene production, emission factors may vary from < 0.1 to 6 kg CH<sub>4</sub>/t. There is not enough data to change the emission factor used in Finland, because it seems that emission depend largely on plant type. Measurement data would provide a more accurate emission factor. However, the share of this emission source of Finnish total GHG emissions is minor (< 0.01%).

## 4.3 Coke production (CRF 2C1)

For CH<sub>4</sub> emissions from coke production, Finland uses an IPCC (1997) emission factor of 0.5 kg CH<sub>4</sub>/t coke. Emission measurements performed in Finland reveal that emission factor could be much lower than the IPCC default used, i.e. 0.1 kg CH<sub>4</sub>/t coke (Pipatti, 2001; Hemminki, 2000). In the National Inventory Report of the UK (Baggott et al., 2004), even lower emission factor is used (0.081 kg CH<sub>4</sub>/t coke). These results reveal that the emission factor used in Finland may overestimate emissions, and the use of an emission factor of 0.1 kg CH<sub>4</sub>/t coke could be justified based on the measurements carried out in Finland as well as the EF used in the UK. This emission source has only a minor importance on the Finnish greenhouse gas emissions (0.01% in 2002).

#### 4.4 New industrial sources

The IPCC is preparing new guidelines for greenhouse gas inventories that will be published in 2006. During the work, some potential new industrial greenhouse gas emission sources are identified. Most new emission sources are related to fluorinated gases, but also some possible sources of  $CO_2$ ,  $CH_4$  and  $N_2O$  have been identified. Below, a preliminary list of possible new emission sources that may occur in Finland is presented:

- copper production
- zinc production
- glass production (CO<sub>2</sub>)
- brick manufacturing (CO<sub>2</sub>)
- mineral wool production (CO<sub>2</sub>)
- ceramics (CO<sub>2</sub>)
- titanium oxide production (CO<sub>2</sub>)
- petrochemical processes (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O).

Emission factors are not yet available for these sources, and work is underway to examine if they are significant enough to be included in emission reporting. In addition, some sources may already be included in reporting (e.g. under soda ash use).

In addition, IPIECA (International Petroleum Industry Environmental Conservation Association) has published guidelines for estimation of greenhouse gas emissions from petroleum industry (IPIECA, 2003). The guidelines give emission factors for different activities in petroleum industry, some of which may also occur in Finland.

## 5. Discussion and conclusions

Finland is obligated to report human-induced greenhouse gas emissions annually to the UNFCCC. Methodologies and emission factors used in greenhouse gas inventories have to be well based and documented. The aim of this report was to evaluate currently used emission factors (mainly  $CH_4$  and  $N_2O$ , and to a lesser extent, CO and NMVOC) for stationary combustion and industrial processes, and to give recommendations for further improvements of the inventory.

The study was carried out using literature survey, available measurement data and expert interviews. It was challenging to compare emission factors from different sources because of, e.g., different classifications and definitions used in literature. It is evident that many variables have an effect on the  $CH_4$  and  $N_2O$  emission factors. Emission factors should be different for, e.g., different combustion technologies, fuel mixes and unit sizes. Other factors, for example load and air ratio, have also a major effect on the emission factors in some cases.

It was also noted that most measurements, of which data are publicly available, were done with full load and in optimal conditions. Therefore, these measurements do not represent average annual emissions.

Main reasons for high methane emissions are small air ratio, short residence time in furnace, low temperature and poor mixing. Too small air ratio or other disturbances occur rather rarely in most cases but their effect on instantaneous emissions is large. This causes uncertainty to emission estimates that are based on calculations. In addition, there are plants which are started up several times a year. For these plants, emissions from start-ups and disorders may play a significant role in the annual emissions. Therefore, measurements done in modern and large plants are not likely to be representative for all plants of the same type.

Methane emission factors are typically larger for small combustion units than for large ones. Methane emissions from natural gas fired engines can be very high, but emissions are likely to be much lower for engines with catalyst. In Finland, use of catalysts is common in gas engines.

Low CO emissions usually indicate low methane emissions. Correspondingly, high CO emissions often indicate high methane emissions in low temperatures. On the other hand, in high temperatures and oxygen-poor atmosphere, methane is decomposed and forms soot but CO is a stable compound. Some large Finnish power plants have continuous CO measurement equipment. This measurement data could probably be used for estimating  $CH_4$  emissions.

Temperature in the furnace is the most important factor affecting N<sub>2</sub>O emissions in addition to fuel used. In fluidized bed combustion (especially in CFB), temperature is lower than in other combustion technologies and therefore N<sub>2</sub>O emissions are higher. Formation of N<sub>2</sub>O depends also on the operating conditions, fraction of excess air and catalytic activity of char and mineral matter. Because formation temperature of N<sub>2</sub>O is low, N2O emissions derive mainly from fuel nitrogen, and emissions are higher for coal than for peat, and higher for peat than for wood. Partial load that occurs most often in district heating plants may increase N2O emissions significantly because temperature decreases with decreasing load. This is notable especially in the case of CFB, where combustion temperature is relatively low even in full load. More measurement data is needed to be able to estimate the relationship between load and N<sub>2</sub>O emissions. In addition, to be able to give representative average emission factors for Finnish plants, more information is needed on typical loading conditions in different types of plants. Combustion temperature may also be low due to combustion of moist fuels, but more information on the effect of fuel moisture on temperatures in the furnace for all combustion technologies would be needed for conclusions.

Results of this study revealed many potential areas for improvements in the emission factors used in the inventory. In this report, recommendations to update the  $CH_4$  and  $N_2O$  emission factors from boilers are given including guidance on the level of the calculations. To be able to further improve emission factors, more measurement data is needed especially from co-firing of different fuels, effect of partial load on  $N_2O$  in FBC and  $CH_4$  emissions from small-scale combustion of biomass.

 $CH_4$  and  $N_2O$  emission factors from industrial processes (nitric acid production, ethylene production and coke production) were also concerned in the study. In the case of nitric acid production, emission factor that is currently based on measurements done in 1999, could be changed based on more recent measurement data. However, continuous on-line measurements are planned to begin in the plants, and these measurement results could be directly used in the inventory in the future. For other industrial sources, no basis for change in emission factors was identified when the importance of the emission sources was taken into account.

CO and NMVOC emissions from stationary combustion were also concerned in the study, but to a lesser extent than  $CH_4$  and  $N_2O$ . For NMVOC emissions, it is recommended that non-zero emission factor for plants > 1 MW (for which the currently used emission factor is zero) would be used. CO emissions vary largely between different plants, fuels and process conditions. Because CO emissions were examined in a more superficial way than the  $CH_4$  and  $N_2O$  emissions in this study, data collected from literature were presented, but no recommendations for emission factors were

given. Data from continuous measurements from plants should be used as much as possible when estimating CO emissions.

Under the UNFCCC, greenhouse gas emissions have to be estimated from 1990 onwards. It seems evident, that e.g. fuel mix, process conditions and plant design and maintenance have changed over time. For example, better adjusting of air ratio has possibly decreased the average  $CH_4$  emission factor by magnitude, and changes in fuel properties, plant design, combustion control and maintenance may have led to change in N<sub>2</sub>O emissions according to Finnish experts. In addition, great changes have occurred in NO<sub>x</sub>-reduction devices which may also have affected  $CH_4$  and  $N_2O$  emissions significantly. But, there is not enough quantitative information available to back up these arguments. Therefore, it was recommended to use the same emission factors for the entire time series.

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### Appendix A: $CH_4$ and $N_2O$ emission factors used in the Finnish GHG inventory in 2002

Table A.1.  $CH_4$  emission factors used in the Finnish GHG Inventory in 2002 for point sources [mg  $CH_4/MJ$ ].

Fuel	Technology/Size [MW]	<u>&lt;</u> 1	1–5	5-15	15-50	50-100	100-150	> 150
					mg Cl	H₄/MJ		
Coal	CFB with air staging	35						
Coal	CFB without air staging	5		5		4		
Coal	BFB + flue gas recirculation				5			
	Burners, grate,							
Coal	burner + grate, others		7	4	4	4	4	4
Coal	Grate + flue gas recirculation		8					
Coal 50-80%	CFB with air staging	35						
Coal 50-80%	PFBC, grate			4			4	
Coal 50-80%	Burner							8
Coal 50-80%	No specification			3				
Oil	Cyclone				10			
Oil	BFB		5					
	Other boilers and > 15 MW							
Oil	gas turbines	10	8	8	8	8	8	8
Oil 50-80%	All		8		3	4		4
Peat	Grate, burner + grate	50	5	2		7		
Peat	FBC, gasification, burners, others		5	5	5	7	7	2
Peat 50-80%	Burner		5					2
Peat 50-80%	Burner + grate				9			
Peat 50-80%	Grate		5	2	15	15	15	
Peat 50-80%	BFB			5		2	2	2
Peat 50-80%	BFB + burner						15	
Peat 50-80%	CFB, other boilers		5		5	5		5
Wood/bark	FBC, gasification		30	30	30	30	30	30
Wood/bark	Other boilers	200	50	50	50	30	30	30
Wood/bark 50–80%	Gasification + air staging	200	50	4	50	50	50	50
W 000/ bark 50 00/0	FBC (also 100–150 MW			-				
Wood/bark 50-80%	BFB + burner)		35	35	35	4	4	4
Wood/bark 50-80%	Grate		35	35	35	20	20	
Wood/bark 50-80%	Burner + grate		50	25		20	20	
Wood/bark 50-80%	Burner					4		
Wood/bark 50-80%	Other boilers		50	50	50			
Black liquor	All boilers (also FBC and grate)				1	1	1	1
Gas	Burner + grate				4	4	3	_
Gus	Other boilers and $> 5 \text{ MW}$					•		
Gas	gas turbines	3	3	3	3	3	3	3
Gas 50–80%	Burner			3	3			7
Gas 50-80%	Low NO <sub>x</sub> -burner + grate				15			
Gas 50–80%	Burner + grate			6				7
Gas 50–80%	No specification				1	5		
Hospital waste	All (also gasification)	50	50	8				
No specification, mixed			35		15	10	10	

No specification, mixed	BFB				15	15	15	15
No specification, mixed	Burner	50	35	6	4	6		7
No specification, mixed	CFB with air staging		30					
No specification, mixed	No specification	50	50	15	15	15	15	7
No specification	Diesel/Otto-cycle engine	2	2	2	2	2		
No specification	Gas turbine					3		
No specification	Coking plant, blast furnace					0		
No specification	Sintering plant	4						
No specification	Rolling mill, melting stoves		0					
No specification	Cupola furnace				8-	-10		
No specification	Other emissions				1	0		
No specification	Other combustion *					8		

\* For example lime kiln, asphalt mixing plant, brick kiln, drying oven and drying plant.

Table A.2.  $N_2O$  emission factors [mg  $N_2O/MJ$ ] used in the Finnish GHG emission inventory for point sources in 2002.

Fuel	Technology	mg N <sub>2</sub> O/MJ
Coal	Grate, burner + grate	4
Coal 50-80%	Grate	2
Coal 50–100%	FBC	70
Coal 50-100%	PFBC, burners, other boilers	2
Oil	FBC	30
Oil 50–100%	Other boilers	2
Peat 50-100%	CFB	30
Peat 50-100%	BFB, other boilers	2
Wood/bark 50-100%	CFB	10
Wood/bark 50-100%	BFB, other boilers	2
Black liquor	All boilers (also FBC and grate)	1
Gas 50–100%	Burner + grate	2
Gas 50–100%	Other boilers	1
Hospital waste	All	2
No specification, mixed	CFB	30
No specification, mixed	BFB, other boilers	2
No specification	Diesel/Otto-cycle engine	31
No specification	Gas turbines, combined cycles	1
No specification	Other combustion *	2

\* For example lime kiln, coking plants, blast furnaces, drying ovens, drying plants, sintering plants, rolling mills, melting stoves, asphalt mixing plants, brick kilns and cupola furnaces.

Table A.3.  $CH_4$  and  $N_2O$  emission factors used in the Finnish GHG emission inventory for small-scale combustion in 2002.

Fuel	CH <sub>4</sub> [mg/MJ]	N <sub>2</sub> O [mg/MJ]
Coal	300	4
Oil	10	2
Natural gas	3	1
Peat	50	4
Wood	300	4

Table B.1 presents emission factors from measurements. Measurement results of engines are for steady-state, but in unsteady situations emissions may be higher.

Table B.1. Measurement data from different sources. $CH_4$ , $N_2O$ , $CO$ , $NO_x$ and $SO_2$ emissions are presented as $mg/MJ$ , if not otherwise
stated. Emissions smaller than detection limit of measurement instruments are presented as ' <detection for="" limit'.="" low<="" lsmgo="" stands="" td=""></detection>
sulphur marine gas oil.

surprise musine sus on.	ne sus ou.													
Fuel	Technology	Capacity	Used since	Bed [°C]	Upper furn. [°C]	Load [%]	CH4	N <sub>2</sub> O	C0	O <sub>2, dry</sub> [%]	NOx	$SO_2$	Source Notes	Notes
Peat	CFB	299 MW <sub>th</sub>	1990	865	901	98	1	< 2	14 ppm	6.1	68	153	[1]	Temperature after separator 905 °C
Peat	CFB	185 MW <sub>th</sub>	1996	865	746	75	<1	26	4 ppm	6.5	128	147	[1]	With 65% load 36 mg N <sub>2</sub> O/MJ. After sep. 860 °C
Peat	BFB	$260 \mathrm{MW}_{\mathrm{th}}$	1993	817		96	2	<1	179 ppm	4.9	136	362	[1]	
Peat	Stoker	$\sim 1 \ \text{MW}$		I	820			22	610	11.7	410		[2]	N-content of fuel: 1.79 w-%
Peat + sludge	BFB	$78  \mathrm{MW}_{\mathrm{th}}$	1996	831		74	3	< 2	113 ppm	5.4	136	121	[1]	Share of sludge only few percents
Peat + wood	CFB	97.5 MW <sub>th</sub>	1995	867		96	1	3	34 ppm	5.5	122	93	[1]	Peat 80% + wood 20%. After separator 867 °C
Peat + wood	CFB	299 MW <sub>th</sub>	1990	846	887	66	1	4	9 ppm	6.2	62	113	[1]	Peat 85% + wood 15%. After separator $894 \ ^{\circ}C$
Peat + bark	BFB	18 MW	1996	897		76	< 1	< 2	38 ppm	6.4	124	40	[1]	Peat 50%
Peat + bark + coal	CFB	$> 300 \ \mathrm{MW}_{\mathrm{th}}$		905	850	100	< 1	18	19	3.1	29	90	[3]	Peat > 50%. Emissions without NOx or SOx reduction methods
Bark	CFB	185 MW <sub>th</sub>	1996	838	768	75	1	10	24 ppm	7.1	123	< 8 <	[1]	T after separator 796 °C
Bark	BFB	$115 \mathrm{MW}_{\mathrm{th}}$	1994	853		64	72	< 2	769 ppm	8.9	73	< 9	[1]	Too small air ratio (average O <sub>2</sub> 3%)
Bark/wood	FBC	> 100  MW						9					[4]	
Bark/wood residues	CFB							5					[1]	Original source: Foster Wheeler, Compact CFB
Bark + wood	Grate	3 MW					4						[5]	In two measures fuel contained also 7.5–12.5% straws. Wood is saw dust.
Bark + wood + waste	Grate	3 MW					2	0 ~					[5]	Waste is pelletized waste (20% and 30% of fuel in these measurements). Wood is saw dust.

## Appendix B: CH<sub>4</sub> and N<sub>2</sub>O emission factors from measurements

Bark + wood + waste	BFB	150 MW				$\sim 90$	0~	0~					[5]	Waste is pelletized waste (15% of fuel). Wood is saw dust.
Wood + waste	CFB	9 MW					0~	0~					[5]	Wood chips + pelletized waste.
Wood residues	Stoker	~ 1 MW		I	950			< 2	671	12.8	250		[2]	Saw dust where $\sim 30\%$ chip board saw dust, includes glue but is very dry (results in high combustion temperature and high LHV). N-content of fuel: 0.54 w-%.
Wood residues	Stoker	$\sim 1 \ MW$		I	770			< 2	1890	13.7	115		[2]	Saw dust and cutter chips. N-content of fuel: 0.20 w-%.
Wood + bark + coal + peat	CFB	$> 300 \text{ MW}_{th}$		850	850	100	~	S	10	4.2	73	v	[3]	Wood and bark cover > 70% of fuel mix. Wood is logging residues.
Wood/bark	BFB	$70~\mathrm{MW}_\mathrm{th}$	1996			52- 56	0.5	1.1	10–18	6.5–6.9	51-48	5.8-5.4	[6]	Softwood saw dust 45% (N-content 0.06 w-%), rotten softwood chips 55% (N-content 0.05 w-%).
Wood/bark	BFB	70 MW <sub>th</sub>	1996			60– 64	1.0	1	23–31	6.3–6.0	91-105	2.5-6.6	[9]	Softw. saw dust 20% (N-content 0.06 w-%), rotten softw. chips 24% (N-content 0.05 w-%), brown chips 56% (N-content 0.5 w-%)
Wood/bark	BFB	70 MW <sub>th</sub>	1996			62	0.5	1	21	9	121	7.8	[6]	Softw. saw dust 20% (N-content 0.06 w-%), rotten softw. chips 24% (N-content 0.05 w-%), green chips 56% (N-content 0.5–0.6 w-%)
Wood/bark	BFB	$70~\mathrm{MW}_\mathrm{th}$	1996			63	0.5	1	14	6.3	143	9.5	[9]	Softw. saw dust 22% (N-content 0.06 w-%), rotten softw.chips 26% (N-content 0.05 w-%), green chips 52% (N-content 0.5–0.6 w-%)
Biofuel	CFB	196 MW <sub>th</sub>				75		20					[1]	Original source [8]. Load is from source [3]
Black liquor	Rec. boiler	2600 tds/d	1990		1038	98	1	<1	195 ppm	4.4	86	< 6	[1]	Liquor from soft wood
Black liquor	Rec. boiler	2600 tds/d	1990			95	<1	<1	25 ppm	4.3	79	< 6	[1]	Liquor from hard wood
Black liquor	Rec. boiler	3000 tds/d	1996		1068	104	<1	<1	5 ppm	3.4	60	< 6	Ξ	Liquor from soft wood
Pulverised coal	Tangential-	430 MW.	1984	I		105	<1	<1	17 ppm	5.9	224	74	[1]	OFA off
	firing					201	<1	<1	17 ppm	5.9	156	66	[1]	OFA on
Pulverised coal	Boxer-firing	$1300 \mathrm{MW}_{\mathrm{evol}}$	1993	I		100		$\sim 1$	1 ppm	4.4	138		[1]	Before SCR. Low-NO <sub>x</sub> burners, OFA
		1201	222			22		<	1 ppm	4.4	59		[1]	After SCR. Low-NO <sub>x</sub> burners, OFA

Pulverised coal	L	1111 070				2	~	1.0	6.5	6.9	154	67	[7]	In the stack, N-content of fuel 1.3 w-%.
	۲r	land W IVI VOC	19/4	I	~ 300	<i>4</i> 0	< 1	1.7	7.8		171	582	[7]	Before FGD, N-content of fuel 1.3 w-%
Dulvarised coal	DF	MM 092	1074		000	03	<1	0.8	4.2	7.3	153	74	[7]	In the stack. N-content of fuel 1.3 w-%.
r uiveiliseu cuai	I.I.	land vy fuel	17/4	I		<i></i>	<1	1.0	3.8	7.6	181	571	[7]	Before FGD. N-content of fuel 1.3 w-%
Dulvarisad coal	DE	360 MW	1077		000 ~~	80	< 1	0.8	3.5	7.6	160	72	[7]	In the stack. N-content of fuel 1.3 w-%.
	1.1	lauf vy fuel	F//1			07	< 1	1.0	4.3	7.8	185	580	[7]	Before FGD. N-content of fuel 1.3 w-%
Dulverised coal	рF	360 MW.	1974	I	о <del>00</del> ~	88	<1	0.6	3.6	7.7	171	57	[7]	In the stack. N-content of fuel 1.3 w-%.
	11	lauf we fuel				00	<1	1.0	4.5	8.3	195	612	[7]	Before FGD. N-content of fuel 1.3 w-%
Hard coal	FBC	< 100  MW						37					[4]	
Coal	PFBC	$\sim 180~\mathrm{MW}$		850	867	103		8	1.1	4.9	8	5	[7]	Pressure in combustion zone 10-11 bar
Coal	PFBC	$\sim 180~{ m MW}$		850	873	102		8	1.1	5.0	6	5	[7]	Pressure in combustion zone 10–11 bar
Coal	PFBC	$\sim 180~{ m MW}$		850	876	98		8	1.2	5.0	10	5	[7]	Pressure in combustion zone 10-11 bar
Coal	PFBC	$\sim 180~\mathrm{MW}$		850	861	97			0.0	5.9	17	3	[7]	Pressure in combustion zone 10-11 bar
Coal	PFBC	$\sim 180~{ m MW}$		849	860	97			0.2	5.9	19	3	[7]	Pressure in combustion zone 10-11 bar
Coal	PFBC	$\sim 180~\mathrm{MW}$		849	860	96		18	0.0	6.0	21	2	[2]	Pressure in combustion zone 10-11 bar
Coal	PFBC	$\sim 180~MW$		850	861	94		10	0.0	6.0	22	3	[7]	Pressure in combustion zone 10-11 bar
Coal	PFBC	$\sim 180~{ m MW}$		850	860	95		12	0.2	5.4	16	7	[7]	Pressure in combustion zone 10-11 bar
Coal	CFB	196 MW <sub>th</sub>				75		35					[1]	Original source [8]. Load is from source [3].
Coal	CFB	$> 300 \mathrm{MW}_{\mathrm{th}}$		905	870	100	<1	30	32	5.1	76	135	[3]	Emissions without NO <sub>x</sub> or SO <sub>x</sub> reduction methods
Lignite	FBC	> 100 MW						14					[4]	
Natural gas	GTCC	$327 \mathrm{MW}_{\mathrm{th}}$	1991	Ι		47	<1	< 3	7 ppm	14.7	70		[1]	Two turbines + one steam generator
Natural gas	GTCC	$113 \mathrm{MW}_{\mathrm{th}}$	1993	Ι		100	<1	< 3	64 ppm	14.9	67		[1]	
														Supplementary firing 20 MW; natural gas did not mix well with the coming
Natural gas	GTCC	$113 \mathrm{MW}_{\mathrm{th}}$	1993	I		115	26	~ ~	122 ppm	13.9	48		[1]	oxygen in supplementary firing
Heavy oil	Boiler	$65 \mathrm{MW}_{\mathrm{th}}$	1983	I		50	<1	<1	8 ppm	4.6	135	387	[1]	
Heavy oil	Boiler	$65 \mathrm{MW}_{\mathrm{th}}$	1983	I		73	<1	<1	9 ppm	4	139	385	[1]	
LSMGO	Gas turbine	2 * 17.2 MW		I		87	1	9	7	17.5	1310	43	[6]	N-content of fuel: 0.005 w-%
LSMGO	Diesel engine	6.9 MW		Ι		90	<1	4	49	12.3	474	32	[6]	N-content of fuel: 0.006 w-%.
LSMGO	Diesel engine	7.1 MW		I		92	<1	1	89	12.7	157	19	[6]	SCR, Nitrogen content of fuel was 0.02 w-%.

78         11.1         1590         42         [9]         N-content of fuel: 0.006 w-%.	[9] N-content of fuel: 0.006 w-%.	[9] N-content of fuel: 0.006 w-%.	[9] N-content of fuel: 0.005 w-%	<b>33</b> [9] N-content of fuel: 0.005 w-%	<b>33</b> [9] N-content of fuel: 0.005 w-%	[10] Typical factors from measurements
[6]	[6]	[6]	[6]	[6]	[6]	[10]
42	43	43	33	33	33	
1590	1362	1112		1012		
11.1	11.9 1362	14.6	10.5 1269	12.3	13.4	
78	86	226	50	80	150 13.4 878	2–3 0–2 35–40
7	6	4	3	4	S	0-2
53 <1 7	46 <1 6	1	76 <1 3	1	2	2–3
53	46	26	76	38	17	
I	Ι	Ι	Ι	Ι	Ι	Ι
0.5 MW	0.5 MW	0.5 MW	4 * 0.6 MW	4 * 0.6 MW	4 * 0.6 MW	1.5 MW
Diesel engine 0.5 MW	Diesel engine 0.5 MW	Diesel engine 0.5 MW	Diesel engine 4 * 0.6 MW	Diesel engine 4 * 0.6 MW	Diesel engine 4 * 0.6 MW	Diesel engine 1.5 MW
LSMGO	LSMGO	LSMGO	LSMGO	LSMGO	LSMGO	Heavy oil

Sources, footnotes and measurement techniques:

[1] Korhonen et al., 2001; Fabritius et al., 2002. Measurements done using NDIR for N<sub>2</sub>O and FID for TOC. Emission factors for methane are measured as TOC. More information about measurement methods used in this source is available in Section 2.1.5.

[2] Linna, 1992. Measurement technique: FTIR.

[3] Roppo, 2004. Measurement technique: FTIR for N<sub>2</sub>O and CH<sub>4</sub>. Other emissions and oxygen measured also using other methods.

[4] Orthofer et al., 1996.

[5] Calculated from measurement results of Vesterinen, 2004.

[6] Harju, 2001. Measurement technique: FTIR.

[7] VTT Chemical Technology, 2000. Measurements of PF plant were done using FTIR technique. NO<sub>x</sub> is calculated by formula NO<sub>x</sub> = NO<sub>2</sub> [mg/MJ] + NO [mg/MJ].

[8] Rautanen et al., 1997.

[9] Cooper, 2001. Measurements were done using Carlo-Erber gas chromatogram (GC) equipped with an electron capture detector, a flame ionisation detector and a thermocouple detector. More information about measurement methods used in this source is available in Chapter 2.1.5.

[10] Kytö, 2004.

## **Appendix C: Emission factors from other countries**

This Appendix presents emission factors from different countries with which emission factors used in Finland are compared. Emission factors from following countries are collected: Austria (Radunsky et al., 2003), Sweden (Feldhusen et al., 2004), Ireland (McGettigan & Duffy, 2003), USA (EPA, 2004), The Netherlands (Spakman et al., 2003), UK (NAEI, 2004), Belgium (Belgium Ministry for Social Affairs, Health and Environment, 2002), Germany (Rentz et al., 2002; Pfeiffer et al., 2000) and Denmark (Denmark, 2004). Because of different classification used in different countries, some simplifications and generalisations were done when preparing the tables.

	Incloud	UK	Sweden		Belgium		USA	A
	Ireland	UK	Sweden	Flemish	Walloon	Brussels	USA	Austria
Public power plants								
Peat	0		20					
Coal	0	0.8	2	0.5	0.2		1	5
Lignite, brown coal								7
Fuel oil, gasoil	0-2	0.1-1	1–2	0-0.6	3.5		3	1–2
Natural gas	0		1	0.1-1.8	0.07		1	1.5
LPG, other gases	0–2		1	0.3-0.5			3	1.5
MSW		0.1	20		0.2			
<b>Residential and commercial</b>								
Peat, peat briquettes	50		30					
Coal, lignite, brown coal	100		4	10.2		200	300	90–230
Brown coal briquettes								7-100
Coke, petroleum coke	50		4					16–90
Fuel oil, gasoil, kerosene	0-5		2–3	2.9		7	10	0.5-2
Natural gas	5		1	1		5	5	0.8
LPG	0		1					1.5
Biomass	30		250			300	300	112-170
Industry								
Coal	15		2	0.31		200	10	5
Coke, petroleum coke	5	0.7	2					2
Fuel oil, gasoil, kerosene	0-2		1–3	0.1		1–3	2	0.1-2
Natural gas, LPG	2		1	0.33		2.5	5	1.5
Scrap tyres		30.4						
Black liquor			0					2
Tall oil			2					0.1
Dry sludge				0				1.5
Biomass	30		30				30	2
Agriculture								
Stationary gasoil	5		2			7		0.5
Biomass	30		250			300	300	170
Agricultural waste								0.7

Table C.1. CH<sub>4</sub> implied emission factors [mg/MJ] for stationary combustion used in Ireland, UK, Sweden, Belgium, USA and Austria.

Technology	Fuel	CH <sub>4</sub>	СО	$N_2O$
All	Steam coal	1.5-15	10	3
All	Petroleum coke	15	61-1000	3
All	Kerosene	7	20	2
All	Gasoil, LPG	1–2	15-100	2
All	Residual oil, orimulsion	3	15-100	2
Boilers < 300 MW	Biogas	4	36	2
Stationary engines	Biogas	323	273	0.5
Gas turbines, boilers < 50 MW	Refinery gas	2	15	2
Boilers $\geq$ 50 MW	Natural gas	6	15-28	1
Boilers < 50 MW	Natural gas	15	15-28	1
Stationary engines	Natural gas	520	175	1.3
Gas turbines	Natural gas	1.5	6.2	2
Public power, CHP, distric	t heating and industrial combustion			
All plants of category	Municipal wastes	0.6	< 8	< 1.3
All plants of category	Wood and similar	0-32	50-240	0–4
Boilers < 300 MW	Agricultural waste, straw, fish & rape oil	0.5-32	63-325	1.4-4
Stationary engines	Fish & rape oil	200	325	4
Commercial, institutional,	forestry, agri- and aquaculture			
All plants of category	Municipal wastes	6	10	4
All plants of category	Wood and similar	200	240	4
All plants of category	Agricultural waste	200	325	4
Residential				
All plants of category	Wood and similar	200	9000	4
All plants of category	Agricultural waste	200	4000	4

Table C.2. CO,  $CH_4$  and  $N_2O$  emission factors [mg/MJ] used in Denmark for energy, manufacturing and transformation industry and non-industrial combustion.

Table C.3. Emission factors for small-scale combustion used in Germany (Pfeiffer et al., 2000). In Germany, a variety of emission factors are used, but this table gives an overview of the typical EFs.

Fuel	CH4 [1	mg/MJ]	N <sub>2</sub> O [1	mg/MJ]
i uci	Households	Other small consumers	Households	Other small consumers
High rank coals and products	273	9.2	11	4.9
Coke from high rank coals	7.1	20	0.8	0.81
Brown coal briquettes	105	248	4.4	0.47
Natural wood	123	96	1.5	0.99
Distillate oil	0.06	0.02	0.61	0.56
Natural gas	1.1	0.12	0.31	0.34

Table C.4.  $N_2O$  emission factors [mg/MJ] for stationary combustion used in the German GHG inventory.

Fuel	Technology	Size	EF
Coal	DBB, WBB, PC	> 50 MW	4
Coal	FBC	50–300 MW	20
Brown coal	DBB, PC	> 50 MW	3.2–3.5
Brown coal	FBC	50–300 MW	8
Oil		> 50 MW	1
Natural gas		> 100 MW	0.5

Table C.5. N<sub>2</sub>O emission factors [mg/MJ] for stationary combustion used in Ireland, UK, Sweden, Netherlands, Belgium, USA and Austria.

	Inclosed	ШZ	Sweden	Netherlands		Belgium		USA	A
	Ireland	UK	Sweden	Netherlands	Flemish	Walloon	Brusse	els USA	Austria
Public power plants and in	dustry								
Peat	12		10						
Coal, coke, petroleum coke, cement coal	5-14	2–3	20	1.4	1.4–14.1	0.9	12	1.4	1.4
All fuel oils, gasoil, kerosene	10–14	0.6	2–5	0.6	0.6–13.4		13	0.6	0.6–2
Tall oil			5						
Natural gas, refinery gas, LPG	3		2	0.1	0.1-3.2	0.2	1.5	0.1–0.6	0.1–0.6
Other gases	0		2	0.1	0.1-2.8				1
Biomass	4		5	1.4				4	4
Black liquor			5						1.4
Dry sludge					0.6				1
MSW		2.1	5	1.4		4.3		3.1	
Scrap tyres		0.6							
Industrial waste									1.4
Residential, commercial an	d agricult	ure							
Peat, peat briquettes	5		10						
Coal, coke, petr. coke	12		20	1.4	12.2		12	1.4	2
Lignite, brown coal									4
Fueloil, gasoil, kerosene	10		2	0.6	11.5-13.4	4	13	0.6	1
Natural gas, LPG	2		2	0.1	2		2	0.1–0.6	0.1-1
Biomass	4		5	1.4			4	4	3–7
Agricultural waste									7

## Appendix D: CO from stationary combustion

CO emissions occur due to incomplete combustion. For example in grates, which are still rather common in small-scale combustion of solid fuels in Finland, poor mixing of gases, short residence time and low combustion temperature may lead to increasing CO emissions. Especially combustion of moist fuels like wastes and peat may lead to high emissions. CO formation increases at start-ups, disorders, shutdowns and partial load (Raiko et al., 2002).

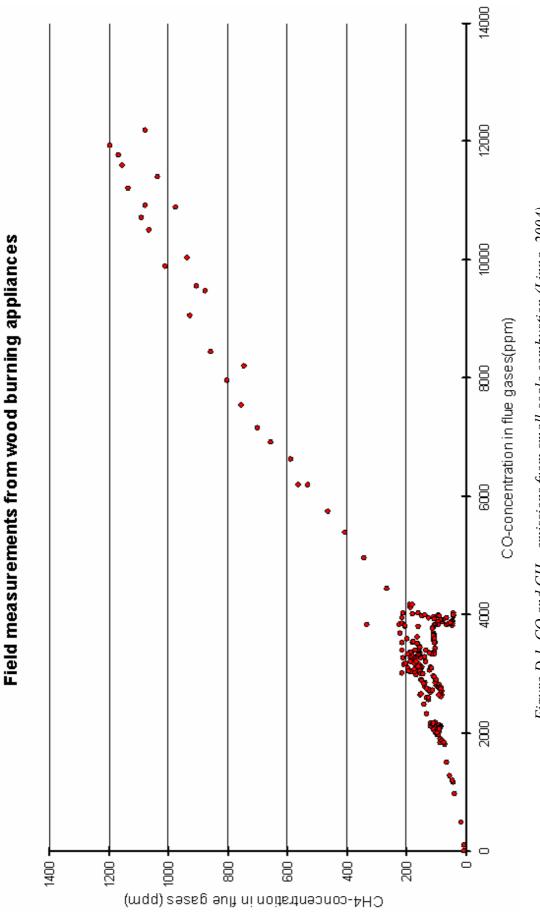
Because carbon monoxide and methane are unburned gases, many properties and formation conditions of carbon monoxide are similar with methane. Some NO<sub>x</sub> reducing technologies like air staging systems and Low-NO<sub>x</sub>-burners increase CO emissions. In the worst cases, CO emissions can increase even tenfold with boiler retrofit for lower NO<sub>x</sub> emissions. There is also opposite information available in the literature. According to the IPCC (1997), effect of emission control methods on CO emissions is negligible for all methods except SCR. For SCR, CO emission reduction of 8% is reported (IPCC, 1997).

Continuous CO measurement equipment are available in some large Finnish power plants (> 50 MW). Measured plant-specific data provides most accurate estimates of the emissions as CO emissions vary much even between plants of the same size and using the same technology. This is due to the large dependency of emissions on load, fuel bit size, fuel moisture, etc. Therefore, measured data should always be used if possible (SYKE, 2004).

In small-scale combustion, variation of emission factors in literature is even larger than in large-scale combustion. Because emissions depend strongly on combustion technology and conditions, country-specific factors should be used. More information about emissions from small-scale combustion in Finland will be available in 2005 when the results of a measurement project (to be carried out at VTT) will be available.

Low CO emissions indicate low methane emissions as the following Figure D.1 and measurement data presented in Appendix B show. But, high CO emissions do not necessarily indicate high methane emissions because high temperature and other conditions may reduce methane emissions even though CO emissions remain high.

For decades, most common technique to measure CO concentrations in flue gases has been non-dispersive infrared (NDIR)-technique. EPA Method 10 is based on NDIRtechnique and European Committee for Standardisation, CEN, is also developing a reference measurement method standard for carbon monoxide measurements, which will be based on NDIR. Nowadays FTIR-technique has also captured the market.





#### D.1 Emission factors for carbon monoxide (CO)

Emission factors for carbon monoxide vary largely between different plants. This Chapter gives a short overview on CO emission factors, but no further analysis or recommendations for emission factors are given. In this Chapter, emission factors used in the Finnish GHG inventory as well as emission factors from literature are presented. More data about measured CO emissions are presented in Appendix B.

#### D.1.1 Coal

Table D.1 presents CO emission factors that are currently used in the national greenhouse gas inventory in Finland for coal combustion. For coke ovens and blast furnaces, emission factor of 0 mg/MJ is used. Table D.2 presents emission factors from literature for refinery gas, coke oven gas and blast furnace gas, and Table D.3 for different coal types.

Fuel	Technology/Size [MW]	< 5	5–15	15-50	50-150	> 150
Coal	CFB with air staging	350				
Coal	Other FBC	100	100	100	30	
Coal	Burner	50		10	10	5
Coal	Burner + grate			30	10	
Coal	Grate	50	30	30	30	
Coal	Grate + flue gas recirculation	20				
Coal	No specification			10	10	10
Coal 50-80%	CFB with air staging	350				
Coal 50-80%	PFBC		20			
Coal 50-80%	Burner and flue gas recirculation					20
Coal 50-80%	Grate				10	
Coal 50-80%	No specification		30			

Table D.1. CO emission factors used in national GHG inventory of Finland [mg/MJ].

Table D.2. CO emission factors [mg/MJ] for refinery gas, blast furnace gas and coke oven gas (Corinair, 2004).

Technology/fuel	Refinery gas	Coke oven gas / Blast furnace gas
Engines > 0.5 MW	10	_
Gas turbines < 50 MW	2	13
All > 50 MW	2–15	0-130 <sup>1</sup>

<sup>1</sup>Corinair90 data.

	)dI	IPCC, 1997				Corinair, 2004	2004		S	<b>SYKE, 2004</b>	1	Measured
Size [MW]					> 50		< 50		15-150	> 150	50	
Technology/fuel	Bituminous/ sub-bituminous	Anthracite	Lignite	Coal 1	Lignite	Coke <sup>(1</sup>	Coal	Coke	Coal > 50%	Coal 50-80%	Coal >80%	CoalCoalCoal50-80%>80%
Pulverized firing	6	310	45	10 - 14	13-16	102-121	73 (other		30	10	5	3.5-7.8
Cyclone furnace	6			10 - 14	10-14 13-16	(coke oven	industrial		30	10		
Fluidized beds	310	5.2	2.8	10 - 14	13-16	coke)	combustion)		30	10	5	0.0-1.2 <sup>(2)</sup>
Utility/industrial stoker	87–190	10		$10^{-1}$	115-160	15	100-196		30	10	5	
				121		(petroleum						
Industrial hand-fed units	4800					coke)				1		
Commercial scale boilers	200			195	195		500-4800	1000	30	-		
Residential combustion	18 (hot	18 (hot water heaters)					4300–7000	1800				
	480–3600 (i	480-3600 (furnaces - stoves)	'es)				(briquettes)	+000		I		

Table D.3. Emission factors from literature and measurements for different coal types [mgco/MJ].

<sup>1</sup> Presented in Corinair (2004) as Corinair90 data.

<sup>2</sup> Pressurised fluidized bed.

Table D.4. CO emission factors for oil combustion from literature and measurements [mgco/MJ].

	)dI	<b>IPCC</b> , 1997	77					Corinair, 2004	2004			SYKE, 2004 Measured	Measured
Size [MW]						> 50				< 50		> 15	
Technology	RFO	DFO	LPG	RFO	Gasoil	LPG	Naphtha	Gasoil LPG Naphtha Kerosene RFO DFO	RFO	DFO	LPG	Oil	Appenuix b
Industrial/utility boilers	$15^{(1)}$	16	16-17	15 <sup>(1</sup> 16 16–17 15	12-15				10	10		20	(mqq 9–8)
Large diesel engines	0.3–350 <sup>(2</sup>	:0 (2		100	12-1130	10–13	5.1	5 <b>C</b>	12–438 12– 601	12– 601	3.3–250 (No technical		35–226
Gas turbines		21		10–15	10–21	(3		12 %	10–30 10–	100	specification)		L
						_				123			
Commercial and residential 5–15 <sup>1)</sup> 16 8–12 17	5-15 <sup>1)</sup>	16	8-12	17	16				13	41-43	13 41–43 10–53		

RFO = residual fuel oil, DFO = distillate fuel oil.

<sup>1</sup> Also for waste oil and shale oil. <sup>2</sup> Small factors are for industrial engines. <sup>3</sup> Presented in Corinair (2004) as Corinair90 data without technical specification.

#### D.1.2 Oil products

Table D.5 presents CO emission factors for oil combustion used in the GHG inventory of Finland, and Table D.4 emission factors from literature.

Table D.5. CO emission factors used in national GHG inventory of Finland for oil combustion [mg/MJ].

Fuel	Technology/Size [MW]	< 5	5-50	50-500
Oil	BFB	100		
Oil	Other boilers and > 15 MW gas turbines	20	20	20
Oil	Diesel engine	46	46	46
Oil 50-80%	All technologies	20	30	10

Measurement results from ships may give a good basis for emission estimates of stationary engines also. Measurement results show that CO emissions vary significantly between similar engines and emissions during unsteady situations are greater than during steady situations (Cooper, 2001). For reserve power engines, amount of start-ups and shutdowns (unsteady situations) might be significant and should be taken into account when defining average emission factors.

In addition, one locomotive engine was measured before and after service. Emission factor for CO was 466 mg/MJ before service and 228 mg/MJ after it. Therefore, service may also have a great effect on CO emissions of power engines (Korhonen & Määttänen, 1999).

#### **D.1.3 Natural gas**

Table D.6 presents emission factors used in the GHG inventory of Finland for natural gas combustion. Table D.7 presents emission factors from literature.

Table D.6. CO emission factors [mg/MJ] used in national GHG inventory of Finland for gas combustion.

Fuel	Technology/Size [MW]	< 15	15-100	> 100
Gas	Burner + grate		70	20
Gas	Other boilers and $> 5$ MW gas turbines	20	20	20
Gas	Diesel engine	46	46	
Gas 50-80%	Burner	200	20	20
Gas 50-80%	Burner + grate	100	100	70
Gas 50-80%	No specification	500	500	500

Source	IPCC, 1997	SYKE, 2004	Corina	ir, 2004
Technology/Size[MW]	_	> 15	> 50 MW	< 50 MW
Utility boilers	18	20	19	
Industrial boilers	16–18		13-17	10
Commercial boilers	9.4		9.6	
Gas turbines	46	20		8-123 (1
Large engines (> 0.5 MW)	340		10-32	2.4-335
Small consumers		20		41
Residential heaters	10			25-250
Residential furnaces	18			25-250
Heavy duty industrial compressor engines				
– Turbines	2			
- 2-cycle lean burn	4.7			
– 4-cycle lean burn	5.1			
– 4-cycle rich burn	20			

Table D.7. CO emission factors from literature for natural gas combustion [mg/MJ].

<sup>1</sup> In one gas turbine measured, EF was 10 mg/MJ at full load but 76 mg/MJ with 50% load.

In Appendix B, results from measurements are presented. In addition, in VAHTI database, measured emission factor of 56 mg/MJ was presented for < 100 MW gas turbine. In Appendix B, no emission factors for CO from gas combustion are given in unit mg/MJ, but there are some measurements results from natural gas firing in combined cycle power plants in unit ppm. Measurements show a large variation in emissions, i.e. from 7 to 122 ppm.

Corinair (2004) presented a comparison of CO emission factors from gas turbines with different  $NO_x$  control technologies (Table D.8), and comparison between old and modern gas turbines (Table D.9). Variation in CO emissions seems to be too large to be able see the difference between different  $NO_x$ -control technologies. But, age of plant seems to be an important factor. CO emission factors from the measurements varied from 1 to 120 mg/MJ, but average for old plants (1970–1980) was about 33 and for new plants about 6 mg/MJ.

Description of turbines	NO <sub>x</sub> control	No of turbines in survey	Size [MW]	NO <sub>x</sub> <sup>(1</sup> [mg/MJ]	CO [mg/MJ]	VOC <sup>(2</sup> [mg/MJ]	N <sub>2</sub> O [mg/MJ]
Public power	Low NO <sub>x</sub> burner	4	150-215	33-86	1.2–21	0.6-2.6	1–7
	Steam injection	1	125	120	4	2.6	< 9
			~ 25-				
	Uncontrolled	2	125	198–250	6.6–51	1.1-4.2	< 9
Combustion	Water injection	1	22	115	20.8	2.8	2
in industry	Steam injection	2	4.8-40	46–91	3–47	1.3-6.3	2
	Uncontrolled	4	6.25-40	140–279	< 3–14	1.6-3.0	< 4-4
Gas extraction / distribution	Uncontrolled	5	6.25–25	104–312	6–120	3.7–25	2–6

Table D.8. Measurements of gas turbines with different  $NO_x$ -control technologies (Corinair, 2004).

<sup>1</sup> Expressed as NO<sub>2</sub>.

<sup>2</sup> Expressed as CH<sub>4</sub>.

Table D.9. Measurements of modern and old gas turbines (Corinair, 2004).

Plant	Capacity	Fuel cons.	CO	NO <sub>x</sub>	Control system	Age of
	MW <sub>th</sub>	GJ/year	mg/MJ	mg/MJ		the plant <sup>1</sup>
Cassanova D'adda 3	105	220,700	32	222	Steam injection	Old
San Donato Milanese 1	12	337,482	34	265	Uncontrolled	Old
San Donato Milanese 2	12	247,068	34	263	Uncontrolled	Old
Turbigo 6	400	1,547,026	_	71	Dry low NO <sub>x</sub>	Medium
Turbigo 7	400	2,336,386	-	77	Dry low NO <sub>x</sub>	Medium
Turbigo 8	400	4,039,245	_	68	Dry low NO <sub>x</sub>	Medium
San Donato Milanese 3	15	392,070	5.9	157	Steam injection	Medium
San Donato Milanese 4	15	343,428	6.1	145	Steam injection	Medium
Sesto San Giovanni	120	3,409,998	16	39	Dry low NO <sub>x</sub>	New
Boffalora	205	1,766,430	5.7	20	Dry low NO <sub>x</sub>	New
Cremona	26	752,536	0.8	108	Steam injection	New

<sup>1</sup>Old: 1970–1980; medium: 1980–1990; new: >1990.

#### **D.1.4 Biomass**

Table D.10 presents CO emission factors for biomass combustion used in the Finnish GHG inventory. Table D.11 presents CO emission factors from literature.

Fuel	Technology/Size [MW]	≤1	1–5	5-50	50-100	100-150	150-500	> 500
Wood/bark	FBC		200	200	200	200	200	
Wood/bark 50-100%	Gasification		0	0				
Wood/bark	Grate, 1–15 MW burners	2100	500	500	200	200	200	
Wood/bark	Grate + burner		500	400				
Wood/bark 50-100%	No specification		500	500	200	200	100	
Wood/bark 50-80%	CFB			500	350	350	350	100
Wood/bark 50-80%	BFB		350	350	350	350		
Wood/bark 50-80%	Burner + grate		500	250	70	200		
Wood/bark 50-80%	Burner				250			
Wood/bark 50-80%	Grate		500	350	75	75		
Black liquor	Recovery boiler			300	200	200	100	100

Table D.10. CO emission factors [mg/MJ] for biomass combustion used in national GHG inventory of Finland.

Table D.11. CO emission factors for biomass combustion from literature. Factors are in mg/MJ if not otherwise stated.

		IPCC, 1997	1997 SYKE, 2004		Corinair, 2004		Measured
Fuel (> 80%)	Technology/ Size [MW]		15-150	> 150	< 50	> 50	Appendix B
Wood	FBC	61	150	150	627 (No	~ 1500	10–31
Wood	Other boilers	290-590	250	150	specif.)	30-300 (2	
Wood	Commercial combustion	440 (1			3600– 7000	199	1890
Wood	Residential comb.	4500– 11000			5790		
Wood pellets	Residential comb.	1700-2300					
Bark	FBC		150	150			80–230 <sup>(3</sup> 24–769 ppm
Bark	Other boilers		250	150			
Black liquor	Recovery boiler					11–314 (2	57 <sup>(4</sup> 5–195 ppm
Biogas	All				7.8–41	13 (2	

<sup>1</sup> If high efficiency.
 <sup>2</sup> Presented in Corinair (2004) as Corinair90 data with no technical specification.
 <sup>3</sup> Boilers capacities around 100 MW. Measured data from VAHTI database.

<sup>4</sup> From VAHTI database, not included in Appendix B.

Factors for woodwaste combustion are presented in Chapter D.1.6 Waste.

#### D.1.5 Peat

Table D.12 presents CO emission factors for peat combustion used in the GHG inventory of Finland. Table D.13 presents emission factors from literature.

Table D.12. CO emission factors used in national GHG inventory of Finland for peat combustion [mg/MJ].

Fuel	Technology/Size [MW]	≤1	1–15	15-50	50-100	100-150	> 150
Peat	CFB		100		100	10	40
Peat	Grate, BFB	200	100	100	40	40	40
Peat	Gasification		0				
Peat	Burners, other boilers		100	100		100	10
Peat 50-80%	Burner		100				40
Peat 50-80%	Burner + grate			70			
Peat 50-80%	BFB, grate		100	40	40	40	40
Peat 50-80%	BFB + burner					70	
Peat 50-80%	CFB, other boilers		100	100	100		100

Table D.13. CO emission factors from literature for peat combustion. Factors are in mg/MJ if not otherwise stated.

	SYKE, 2004		Corinai	r, 2004	Measurements	
Technology/Size [MW]	15-50	50-150	>150	< 50	> 50	See Appendix B
Grate	100	30	10			610
FBC	100	30	10		30–160 (1	4–179 ppm
Other (e.g. PF)	30	10	10			
Residential				18-18533		

<sup>1</sup> Presented in Corinair (2004) as Corinair90 data with no technical specification.

#### D.1.6 Waste

Majority of wastes combusted in Finland are combusted in co-firing with other fuels. Therefore the emission factors correspond with emission factors of the main fuel used in the boiler. For hospital waste incineration, an emission factor of 500 mg CO/MJ is used. Table D.14 presents CO emission factors from literature for waste combustion.

Fuel	IPCC,	Corinair, 2004		Measurements presented in
	1997	< 50 MW	> 50 MW	Appendix B
MSW	14–98	33-2188	19–98	
Agricultural waste		200-8500	20 (1	
Woodwaste		61-8500	12-300 (1	671 (woodwaste on grate)
Industrial waste		15-510		
Waste gas		8.8–27	$0.1 - 25.5^{(1)}$	

*Table D.14. CO emission factors from literature for waste combustion [mg/MJ].* 

<sup>1</sup> Presented in Corinair (2004) as Corinair90 data.

#### D.1.7 Mixed fuels

Table D.15 presents CO emission factors used in the Finnish GHG inventory for combustion of mixed fuels, where share of each fuel is < 50%.

Table D.15. CO emission factors [mg/MJ] used in Finnish GHG inventory for cocombustion.

Technology/Size [MW]	≤1	1–5	5–15	15-50	50-150	> 150
Grate, burner + grate		350		100	50	
Burner	1000	350	100	30	15	25
BFB				20	45	45
CFB with air staging		20				
No specification	1000	500	100	100	50	25

#### D.1.8. Others (e.g. kilns, ovens and dryers)

This category includes combustion in, e.g., lime kiln, asphalt mixing plant, brick kiln, drying oven and drying plant. Emission factors used in the inventory are determined by the process type rather than by fuel. Table D.16 present CO emission factors used in the Finnish GHG inventory.

Table D.16. CO emission factors [mg/MJ] used in the Finnish GHG inventory for combustion, where emission factor is derived by process type rather than by fuel.

Туре	<b>Emission factor</b>
Coking plant, blast furnace, rolling mill, melting stove	0
Sintering plant	1500
Cupola furnace	20–30 (1
Other combustion <sup>(2</sup>	20

<sup>1</sup> 30 for  $\leq$  1 MW, 20 for others.

<sup>&</sup>lt;sup>2</sup> E.g. gas turbines not included to previous tables, lime kiln, asphalt mixing plant, brick kiln, drying oven, drying plant.

Emission factors of IPCC (1997) for kilns, ovens and dryers are presented in Table D.17. For lime kiln, a measured emission factor of 11 mg/MJ was given in the VAHTI database.

Table D.17. CO emission factors for kilns, ovens and dryers [mg/MJ] (IPCC, 1997).

Industry	Emission source	<b>Emission factor</b>
Cement, lime	Kilns – natural gas	83
Cement, lime	Kilns – oil, coal	79
Coking, steel	Coke oven	211
Chemical processes, wood, asphalt, copper, phosphate	Dryer – natural gas	11
Chemical processes, wood, asphalt, copper, phosphate	Dryer – oil	16
Chemical processes, wood, asphalt, copper, phosphate	Dryer – coal	179

## Appendix E: NMVOC from stationary combustion

Emissions of non-methane volatile organic compounds (NMVOC) result from incomplete combustion (low temperature and/or incomplete mixing of fuel and air). Such conditions are rare in large boilers and in general emissions decrease when boiler size increases. In the Finnish GHG inventory, NMVOC emissions from fuel combustion are estimated roughly. The emissions factor for NMVOC emissions for boilers smaller than 1 MW burning wood or bark is 1800 mg/MJ and zero for other boilers. The estimates are based on assumption that in small-scale combustion, share of  $CH_4$  of total VOC is 1/3, and in larger boilers, VOC consists only of methane.

The assumption of negligible emissions from large boilers is based on studies made in the late 1980's and early 1990's, compiled and evaluated by Finnish experts (Boström, 1994; Boström et al., 1992). In this Appendix, firstly an introduction to VOC measurement techniques is given, and secondly, the validity of the simplifying assumption used in the Finnish GHG inventory is assessed.

#### E.1 VOC measurement techniques

There are a number of different definitions of VOC throughout the world which can be based, e.g., upon vapour pressure, boiling point or reactivity of organic species. The most commonly used definition in Europe is the one stated in the EU Solvent Emission Directive, which defines VOC as follows:

*VOC is any organic compound having at 293,15 K a vapour pressure of 0,01 kPa or more, or having a corresponding volatility under particular conditions of use.* 

A number of European countries have, however, developed their own definition in specific contexts. For example, in Austria VOC is defined as *organic compound having a maximum boiling point of 200*  $^{\circ}$ C and in Switzerland VOC is an organic compound having a maximum boiling point of 240  $^{\circ}$ C.

On the other hand, the current definition for VOC according to U.S. EPA and Environment Canada is as follows:

*VOC* means any compound of carbon, excluding carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate, which participates in atmospheric photochemical reactions.

Thus, according to US EPA, e.g. methane and ethane are not to be considered as VOC-compounds.

Existing methods for VOC determination can be divided into following groups:

Continuous measurement methods based on

- flame ionisation detector (FID)
- Infrared Spectroscopy (e.g. FTIR, NDIR).

Non-continuous measurement methods:

- sampling based on
  - absorption
  - adsorption
  - bag sampling
  - sampling in gas vessels
  - dilution sampling.
- analysis based on e.g.:
  - gas chromatography (GC)
  - ion chromatography (IC)
  - high performance liquid chromatography (HPLC)
  - Infrared Spectroscopy (FTIR, IR).

The most common analyzer used for VOC-measurements is *flame ionisation detector (FID)* which is based on the ionization of organically bonded carbon atoms in a hydrogen flame and the discharge of the ions produced to a pair of electrodes. The ionic current, which is measured, is proportional over a wide range to the number of carbon atoms delivered to the flame per unit time. Several standards (e.g. EPA 25A, EN12619, EN13526, VDI 3481) are also based on FID.

The positive attributes for FID method are that sampling and analysis are easy to perform, one can get real time measurement data from the process and the measurements can be performed from hot and wet gases, so there is no need to cool the sample gas.

The main problem with FID method is the variation of the FID response to separate VOC components other than hydrocarbon compounds. For example, VOC compounds containing oxygen (like aldehydes and ketones) or halogen atoms may differ considerably in FID response compared to similar hydrocarbon components.

Also, the response factors are dependent on the detector and on the sample gas matrix, and therefore it is difficult to determine correct response factors for all sampling situations and if this compensation for response factors is not done, the VOC concentrations can easily be either under or over estimated. Since a FID measures the quantity of carbon ions relative to a standard, the individual VOC compounds are not identified. The VOC measurement is a very demanding and challenging task. Without thorough and careful measurement planning, misleading results can be obtained and wrong conclusions can be drawn from these. The selection of a measurement method depends on the purpose of the measurements, whether TOC or separate VOC-compounds need to be determined.

#### E.2 NMVOC emission factors

The following Tables (E.1–E.3) present NMVOC emission factors from different sources by fuel type. In Table E.1 for coal, emission factors for hydrocarbons ( $C_xH_y$ ), which are one group of compounds of NMVOC are also given.

Table E.1. NMVOC emission factors [mg/MJ] for coal combustion and  $C_xH_y$  emission factors for solid fuels from different sources.

IPC	СС, 1997	SYKE, 2004	Corinair, 2004		Maskuniitty, 2002 <sup>1</sup>		
Utility	Industrial		Grate	Boiler	Small boiler	1–5MW	> 50 MW
			> 50 MW	< 50 MW			
5	20	3 <sup>2</sup>	50	600	200–700	< 100 to	< 5 to
						> 300	> 10

<sup>1</sup>C<sub>x</sub>H<sub>y</sub> emission factors based on measurements done at VTT (Juha Huotari).

<sup>2</sup> Based on Corinair and AP-42.

Table E.2. NMVOC emission factors [mg/MJ] for natural gas and oil combustion.

		Corinair,	2004	SYKE, 2004 <sup>1</sup>	IPCC, 1997
Fuel	Boiler	Gas turbine	Stationary engine		
	> 50 MW	< 50 MW	< 50 MW		
Natural gas	5	2.5-5	200	2	5
Oil	5-10	1.5–5	1.5-100	2 (HFO)	5
1				3 (LFO)	

<sup>1</sup> Based on Naturvårdsverket, 2001 and AP-42.

	IPCC, 1997	SYKE, 2004 <sup>1</sup>	Corinair, 2004	
Fuel	<b>Energy industries</b>		Boiler > 50 MW	Boiler < 50 MW
Wood	50	20	80	100-400
Peat		30	30	

<sup>1</sup> Based on Naturvårdsverket, 2001 and Corinair.

For the purposes of this work, we interviewed Finnish experts involved in emission measurements from Finnish power plants (e.g. Jukka Lehtomäki from VTT Technical Research Centre of Finland). Available measurement data did not allow compiling emission factors, but gave a good insight in the variability of emissions. According to the experts, NMVOC emissions depend strongly on operation conditions of the power plant. For fossil fuels NMVOC emissions are often low (< 1 mg/m<sup>3</sup> flue gas) though larger emissions (1–10 mg/m<sup>3</sup>) occur in some conditions. For biofuels, especially for wood combustion, these emissions are significantly larger, from tens to hundreds of mg NMVOC in m<sup>3</sup> flue gas.

According to the values presented in this Chapter, it seems that NMVOC emissions from power plants > 1 MW could also be estimated in Finland. In addition, present EF for < 1 MW plants combusting biomass seems to be too high and classification too rough.



Series title, number and report code of publication

VTT Research Notes 2321 VTT–TIED–2321

Author(s)

Tsupari, Eemeli, Monni, Suvi & Pipatti, Riitta

# Title Non-CO<sub>2</sub> greenhouse gas emissions from boilers and industrial processes

## Evaluation and update of emission factors for the Finnish national greenhouse gas inventory

Abstract

Finland is obligated to report anthropogenic greenhouse gas emissions annually to the UNFCCC. Inventories of greenhouse gas emissions and methodologies used have to be well based and documented. The aim of this report is to give input to Finnish greenhouse gas inventory of energy (stationary combustion) and industrial sectors. The emission factors for the industrial processes used in the inventory are based both on plant-specific factors and the IPCC default values. The currently used emission factors for the non-CO<sub>2</sub> greenhouse gases from combustion are mainly based on studies done at the beginning of 1990's, even though small modifications to the emission calculation model have been done afterwards. Since 1990, significant changes have occurred in plant design and maintenance, operating conditions and composition of fuels. In addition, knowledge of emission generating processes has increased world-wide. In this study, the non-CO<sub>2</sub> emission factors (mainly CH<sub>4</sub> and N<sub>2</sub>O, and at a lesser extent, CO and NMVOC) for combustion and industrial processes used in the Finnish greenhouse gas inventory are evaluated based on domestic and international literature, available measurement data and expert interviews. Results of this study revealed many potential areas for improvements in the inventory. Consequently, recommendations to update the CH<sub>4</sub> and N<sub>2</sub>O emission factors from boilers and industrial processes are given including guidance on the level of the calculations. The time dependency and uncertainties of the emission factors are also considered. In addition, effect of load and fuel mixes on emissions is examined. Areas for further research are also indicated.

Keywords

Activity unit

greenhouse gases, emission factors, energy production, stationary combustion, co-firing, industrial processes, nitrous oxide, volatile organic compounds, methane, carbon monoxide, recommendations, CO,  $CH_4$ ,  $N_2O$ , NMVOC

VTT Processes, Biologinkuja 7, P.O.Box 1602, FI–02044 VTT, Finland						
ISBN 951–38–6759–5 (soft back e 951–38–6760–9 (URL: http:	Project number 35PÄÄSTÖKERT					
Date	Language	Pages	Price			
December 2005	English	82 p. + app. 24 p.	С			
Name of project Evaluation of emission factors for boilers and industrial processes		Commissioned by Statistics Finland				
Series title and ISSN		Sold by				
VTT Tiedotteita – Research	Notes	VTT				
V11 Thedottelta – Research Notes 1235–0605 (soft back edition) 1455–0865 (URL: http://www.vtt.fi/inf/pdf/)		P.O.Box 1000, FI–02044 VTT, Finland Phone internat. +358 20 722 4404 Fax +358 20 722 4374				

This report presents evaluation of non-CO<sub>2</sub> greenhouse gas (GHG) emission factors for stationary combustion and industrial processes used in the Finnish GHG inventory under United Nations Framework Convention on Climate Change. The study focuses on CH<sub>4</sub> and N<sub>2</sub>O emission factors, but CO and NMVOC are also considered. In the report, emission factors from domestic and international literature, measurements and expert interviews are collected and compared with those used in the Finnish GHG inventory. Consequently, recommendations to update the CH<sub>4</sub> and N<sub>2</sub>O emission factors from stationary combustion and industrial processes are given including guidance on the level of the calculations. The time dependency and uncertainties of the emission factors are also considered.

VTT RESEARCH NOTES 2321 Non-CO<sub>2</sub> greenhouse gas emissions from boilers and industrial processes

Tätä julkaisua myy VTT TIETOPALVELU PL 1000 02044 VTT Puh. 020 722 4404 Faksi 020 722 4374 Denna publikation säljs av VTT INFORMATIONSTJÄNST PB 1000 02044 VTT Tel. 020 722 4404 Fax 020 722 4374 This publication is available from VTT INFORMATION SERVICE P.O.Box 1000 FI-02044 VTT, Finland Phone internat. + 358 20 722 4404 Fax + 358 20 722 4374

ISBN 951-38-6759-5 (soft back ed.) ISSN 1235-0605 (soft back ed.) ISBN 951- 38- 6760- 9 (URL: http://www.vtt.fi/inf/pdf/) ISSN 1455- 0865 (URL: http://www.vtt.fi/inf/pdf/)