PROPOSED NO_X EMISSIONS REPORTING FOR HYDROGEN-CONTAINING FUELS

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The current normalisation method for correcting emissions, in particular nitrogen oxides (NO_x), to reference conditions adds a burden of about 37% on hydrogen (H_2) compared to natural gas. This is partly due to (1) the correction to dry flue gas conditions but (2) largely due to the normalisation of the measured values to 15% oxygen (O_2), dry.

1. NO_x and carbon monoxide (CO) are usually measured by extractive sampling methods using probes in the exhaust duct or stack. Traditionally, and as also defined in measurement standards, water (H_2O) is extracted from the sample before gas analysis in order to avoid condensation in the sampling equipment and spectral cross-sensitivities in the analysis, for example. However, burning H_2 at the same thermal input as for methane (CH_4), the main constituent of natural gas, produces more H_2O and no carbon dioxide (CO_2) in the flue gas. Removing this H_2O for the measurement of NO_x, CO and other components in the dry flue gas is equivalent to an increased concentration of the measured component in the dry sample.
2. The normalisation to the reference O_2 content accounts for excess dilution in the combustion process, or leakage in the sampling system, and the normalisation factors are very similar for different hydrocarbons, but significantly different for hydrogen. Consequently, a different normalisation procedure is required to accommodate hydrogen and this should refer to energy specific units rather than flue gas volumes. Equivalent procedures have been proposed by others [1-3].

The differences between this new procedure and the current procedure, which is based on dry O_2 correction alone, is small across the range of hydrocarbon fuels but increases by about 37% for pure hydrogen when relating the NO_x emission to the exhaust volume released per unit of energy as shown in *Figure 1*. *Figure 1* plots the emissions measurement bias imposed when burning a variable volumetric hydrogen content in natural gas (represented here as CH_4).

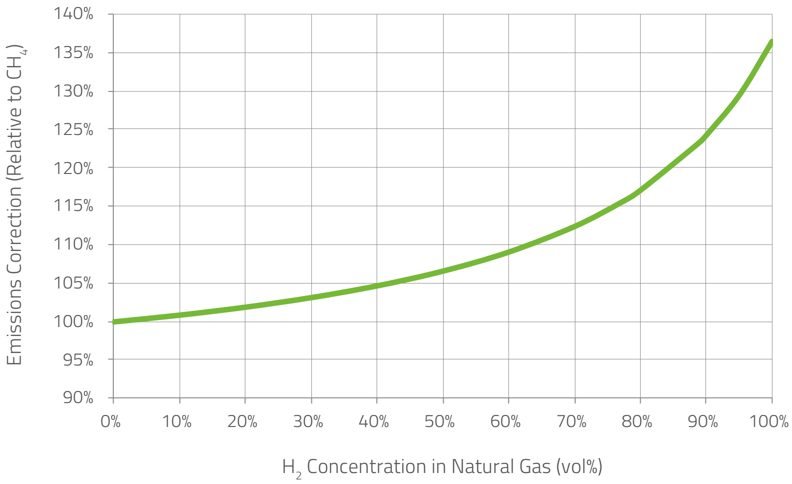

Figure 1. Energy-based emissions correction factor for increasing hydrogen content in natural gas

The following procedure is therefore proposed.

Regulating NO_x emissions from hydrogen gas turbines

In Europe, the mass concentration of NO_x in flue gas is regulated by setting Emission Limit Values (ELVs) in mg/m^3 at a reference condition of 15% O_2, dry, 273.15K, 101.3 kPa. Correction of raw measured NO_x concentrations, in parts per million by volume (ppm), to these reference conditions is performed routinely, using standard formulas, and ensures that dilution of the flue gas by excess combustion air and by water vapor is taken into account [4, 5].

The major constituents of the dry flue gas are CO_2, arising from the oxidation of carbon in the fuel, along with O_2 and nitrogen (N_2) from the combustion air. The flue gas volume, at gas turbine reference conditions, is similar for a range of hydrocarbon fuels, as defined in EN ISO 16911-1 [6] and as shown in *Table 1*. The Fuel Factor is the flue gas volume (m^3) per unit of useful (net) thermal energy (MJ), supplied by the fuel, in m^3/MJ. It can be seen that the Fuel Factor variation is very limited when comparing hydrocarbon fuels. However, since hydrogen is a carbon-free fuel, the flue gas does not contain CO_2 and the resultant dry flue gas volume is substantially lower. Relative to hydrogen, the flue gas volumes from natural gas combustion are therefore about 37% higher on this energy specific basis, as shown in *Table 1*, and therefore natural gas ELVs need to be corrected for hydrogen firing to achieve equivalence.

Table 1. Flue gas volume correction factors for hydrocarbon and hydrogen fuels at gas turbine reference conditions

Fuel	Natural Gas[1]	Gas Oil	Fuel Oil	Hydrogen
Fuel Factor (m^3/MJ) at 15% O_2 dry (unit conversion factor)	0.845	0.859	0.873	0.616
ELV correction factor relative to natural gas	1.000	0.984	0.968	1.372

A given quantity of NO_x (mg), or other pollutants, produced by a gas turbine combustion system, when firing a given quantity of energy, is dispersed within the reference flue gas volume (m^3). The smaller reference flue gas volume for hydrogen therefore gives rise to an artificially high NO_x concentration in relation to the energy released. This can be corrected by regulating NO_x concentration in units of mg/MJ (net thermal input). For gas turbine emissions corrected to 15% O_2 dry conditions, the mass concentration (mg/m^3) can be simply multiplied by the appropriate Fuel Factor (m^3/MJ) in Table 1 to give a NO_x concentration in mg/MJ. The Fuel Factor is a unit conversion factor that transforms mg/m^3 into mg/MJ at a given reference condition, noting that different Fuel Factors are required when firing natural gas or hydrogen in boilers with a reference condition of 3% O_2. The emissions are then comparable so that, when switching from natural gas to hydrogen, the same energy input produces the same NO_x concentration, in mg/MJ, and the same mass release rate from the stack, in mg/s. It should be noted that the formula in the Industrial Emissions Directive [4] for calculating a thermal input weighted average ELV for blended fuels, for Large Combustion Plant, is exactly equivalent to the above approach when the ELVs are in mg/MJ.

Alternatively, the above correction factor given in Table 1 can be used to adjust a natural gas ELV, in mg/m^3 at 15% O_2 dry, to give the equivalent hydrogen ELV, in mg/m^3. The correction factor is independent of the oxygen reference condition and is applicable to both gas turbines and gas fired boilers. To extend this approach to other fuels and fuel blends, a more general scheme is proposed in the Appendix which presents a general equation for deriving the Fuel Factor. This approach is based only on the lower heating value and the reaction stoichiometry of the fuel.

A variable hydrogen content can be accommodated by pro-rating the above unit conversion factors or correction factors using the proportion of hydrogen in the fuel (by energy content).

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[1] Note that the unit conversion factor for natural gas, i.e., the Fuel Factor, is applicable to Group H natural gas (EN 437 [7]) that also conforms to the definition of natural gas within the Industrial Emissions Directive [4]. Conversion factors for other natural gas groups are given in EN 15502-1 [8], for example.

Example

For example, a NO_x ELV of 50 mg/m^3 at 15% O_2, dry, when firing natural gas, is equivalent to 42.25 mg/MJ [= 50 mg/m^3 * 0.845 m^3/MJ]. The same NO_x ELV of 50 mg/m^3, when firing hydrogen, is equivalent to 30.80 mg/MJ [= 50 mg/m^3 * 0.616 m^3/MJ]. This illustrates the penalty that would be applied to hydrogen if the flue gas volume difference is not taken into account.

To achieve parity, an increased NO_x ELV of 68.6 mg/m^3 is required, when firing hydrogen, to give the same result of 42.25 mg/MJ [= 68.6 mg/m^3 * 0.616 m^3/MJ]. In other words, when setting an ELV for pure hydrogen in mg/m^3, this needs to be multiplied by a correction factor of 1.372 to achieve parity with natural gas. An ELV of 50 mg/m^3 for natural gas is then equivalent to an ELV of 68.6 mg/m^3 for hydrogen using the same example.

Conclusions

1. NO_x emissions from the combustion of hydrogen and natural gas are only directly comparable, and equivalent, when the concentrations are reported in mg/MJ (net thermal input). NO_x emissions in mg/m^3 can be multiplied by a Fuel Factor, at the same reference conditions, to obtain mg/MJ. The Fuel Factor is a unit conversion factor. Alternatively, the natural gas ELV in mg/m^3 can be multiplied by a correction factor of 1.372 to obtain the equivalent hydrogen ELV in mg/m^3. Fuel blends can be addressed by energy weighted averaging of the conversion or correction factors.
2. Reporting emissions on an equivalent basis is important so that hydrogen combustion is not penalised and decarbonisation projects are not hindered. However, this does not address the potential need for higher ELVs that may be required when firing hydrogen which depends on the success of ongoing combustion system development [9].

References

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[7] EN 437, Test gases - Test pressures - Appliance categories, 2003.

[8] EN 15502-1, Gas-fired heating boilers - Part 1: General requirements and tests, 2012.

[9] European Turbine Network, Addressing the combustion challenges of hydrogen addition to natural gas: Summary Report, ETN Global Report, November 2022.

Appendix

For a general fuel (possibly composed of several fuel components) of composition $C_nH_mO_lN_pS_k$, the stoichiometric combustion equation (introducing stoichiometric factors) reads like this (per mole of fuel):

$$\left(\frac{m}{4} + n - \frac{l}{2} + k\right) O_2 + C_nH_mO_lN_pS_k \rightarrow n \cdot CO_2 + \frac{m}{2} H_2O + \frac{p}{2} N_2 + k \cdot SO_2$$

with

$$\nu_{stoich} = \frac{m}{4} + n - \frac{l}{2} + k \tag{1}$$

where ν_{stoich} is the stoichiometric air requirement (per mole of fuel). Inerts like N_2 or CO_2 in the fuel flow can be included in the generalised fuel formula.

The lower heating value (LHV) for a mixture of fuels can be averaged and expressed in mass, mole or volume specific terms with a suitable inter-conversion, for example, converting from a mass specific LHV to a volume specific LHV using the fuel density (ρ_{fuel}):

$$LHV\_V \left[\frac{MJ}{m^3}\right] = LHV\_m \cdot \rho_{fuel} \tag{2}$$

The resulting fuel volume per unit of thermal energy is the inverse of the volumetric heating value:

$$V_{fuel} = \frac{1}{LHV\_V} \quad = \quad \left[\frac{m^3}{MJ}\right] \tag{3}$$

The flue gas release (Fuel Factor), S [m^3/MJ], represents the volume of the dry exhaust gas flow after combustion which is comprised of the volumes, V [m^3/MJ], of CO_2, SO_2, O_2 from the excess combustion air, N_2 and other inerts ($N_{2,inerts}$) from the combustion air, and N_2 deriving from the fuel ($N_{2,\ react}$):

$$S = V_{CO2} + V_{O2} + V_{SO2} + V_{(N2,inerts)} + V_{(N2,react)} \tag{4}$$

Air is composed of O_2 (~ 21% by volume) and N_2 along with other inerts (~ 79% by volume), or more generally: the oxygen content of the dry oxidiser (ox) is $x_{O_2,ox,dry} = 0.2095$ and $x_{N_2,inerts} = 1 - x_{O_2,ox,dry}$, where x is the volume fraction. The volume of inerts per volume of air is then:

$$\frac{x_{N_2,inerts}}{x_{O_2,ox,\ dry}} = \frac{1 - x_{O_2,ox,\ dry}}{x_{O_2,ox,\ dry}} = \frac{1}{x_{O_2,ox,\ dry}} \qquad i.e., \sim 3.77 \text{ for dry air} \tag{5}$$

With this, equation (4) can be expressed for stoichiometric conditions $(V_{O2}=0)$ and air as the oxidiser:

$$S_{stoich} = n \cdot V_{fuel} + k \cdot V_{fuel} + \nu_{stoich} \cdot V_{fuel} \cdot \left(\frac{1}{x_{O_2,ox,\ dry}} - 1\right) \frac{p}{2} \cdot V_{fuel} \tag{6}$$

$$S_{stoich} = \frac{n + k + \frac{p}{2} + \nu_{stoich} \cdot \left(\frac{1}{x_{O_2,ox,\ dry}} - 1\right)}{LHV\_V} \tag{7}$$

$$S_{stoich} = \frac{n + k + \frac{p}{2} + \nu_{stoich} \cdot 3.77}{LHV\_V} \tag{8}$$

Correction to 15% O_2 can be done simply:

$$S@15\%O_2 = \frac{n + k + \frac{p}{2} + \nu_{stoich} \cdot 3.77}{LHV\_V} \cdot \left(\frac{0.2095 - 0.0}{0.2095 - 0.15}\right) \quad for\ dry\ air \tag{9}$$

With these equations we can derive and replicate the Fuel Factors in EN ISO 16911-1 for stoichiometric conditions (0% O_2 in the flue gas) and also for the reference condition (15% O_2, dry). For oxidisers other than dry air, containing a different O_2 content ($x_{O_2,ox,dry}$), different relative differences are obtained as expected.

Figure A.1. compares 1) the stoichiometric Fuel Factor for CH_4-H_2 blends, when calculated from first principles using a molar balance approach (*S_direct_stoich*) with 2) the stoichiometric Fuel Factor for natural gas-H_2 blends using the natural gas Fuel Factor defined in EN ISO 16911-1 and an equivalent Fuel Factor for H_2. It can be seen that these values are essentially coincident (*S_ISO 16911-1*). *Figure A.1.* also shows how the Fuel Factor at 15% O_2 (*S@15%O*$_2$) varies with the proportion of H_2 in the blend (by volume).

The ELV correction factor discussed earlier is also plotted as a function of the proportion of H_2 in the blend (Emissions Correction).

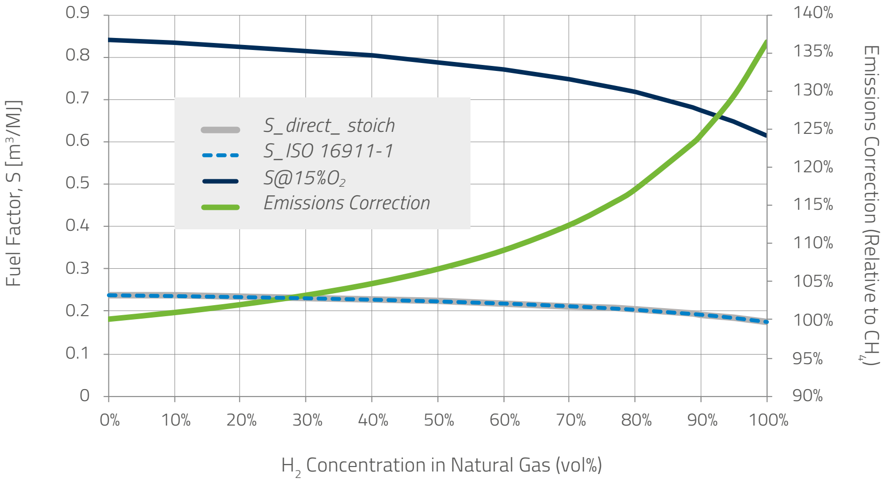

Figure A.1. Fuel Factor and ELV correction factor for natural gas – hydrogen blends

When raw measured ppm is the starting point, the additional steps in the calculation to obtain a concentration in mg/MJ are shown below. If the concentration is measured on a 'wet' basis, the ppm wet needs to be divided by ($1 - H_2O_{,ex}$) to obtain ppm dry, where $H_2O_{,ex}$ is the volume fraction of water vapour in the exhaust (***ex***) gas.

The raw value, expressed as a volume fraction $x_{NOx}\left[\frac{mol}{mol} = \frac{Nm^3}{Nm^3}\right]$ is converted to an energy-related quantity as follows:

Correction to 15% O_2 is performed first where $\boldsymbol{x_{NOx}}$ is given as a volume fraction:

$$x_{NOx,15\%\,O_2} = x_{NOx} \cdot \left(\frac{0.2095 - 0.15}{0.2095 - O_{2\,,ox,\,dry}}\right)\left[\frac{Nm^3}{Nm^3}\right] \tag{10}$$

and $\boldsymbol{O_{2,ex,dry}}$ is the volume fraction of oxygen in the dry exhaust gas.

To give a mass-based emission, multiplication with $\rho_{NO2} = 2.053\left[\frac{kg}{Nm^3}\right]$ yields:

$$m_{NOx,15\%O2}\left[\frac{mg}{Nm^3}\right] = x_{NOx,15\%O2}\left[\frac{Nm^3}{Nm^3}\right] \cdot \rho_{NO2}\left[\frac{kg}{Nm^3}\right] \cdot 10^6\left[\frac{mg}{kg}\right] \tag{11}$$

Since

$$x_{NOx,15\%O2}\left[\frac{Nm^3}{Nm^3}\right] = ppm_{NOx,15\%O2} \cdot 10^{-6}$$

then

$$m_{NOx,15\%O2}\left[\frac{mg}{Nm^3}\right] = ppm_{NOx,15\%O2} \cdot \rho_{NO2}\left[\frac{kg}{Nm^3}\right] \tag{12}$$

Using the Fuel Factor to correct the emissions from ***[mg/m³]*** to energy specific units of ***[mg/MJ]*** (net thermal input) for alternative fuels gives a practical and robust comparison of combustion performance and environmental impact:

$$NO_x = m_{NOx,15\%O2} \cdot S@15\%O_2\left[\frac{mg}{MJ}\right] \tag{13}$$

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