EMISSION PATTERNS OF UNVENTED COMBUSTION DEVICES BURNING LIQUID AND GASEOUS FUELS

V. Haraprasad  J.M. Dave

ABSTRACT

Emission factors of important criteria pollutants like \( \text{NO}_x \), \( \text{SO}_2 \), \( \text{CO} \), particulate matter, and \( \text{HCHO} \) were determined at different burn rates both with and without a pan, in the case of a kerosene wick stove and an LPG stove burning a 55:45 mixture of propane-butane gases. Findings reveal certain patterns in the evolution of these pollutants which are affected by the use of the pan. Also, there appears to exist a range of burn rates wherein the evolution of some pollutants is at an optimum. Non-stoichiometric \( \text{SO}_2 \) emissions, as well as the factors affecting \( \text{CO} \) evolution, are discussed. The paper highlights factors affecting source emissions and outlines practical measures for abatement of emissions at the source.

INTRODUCTION

The source emissions, termed \( S \), in indoor air quality models, are assumed to remain constant and are expressed in volumetric or gravimetric units of the concerned pollutant per unit time (Turk 1963; Moschandreas et al. 1987). However, it has been shown that for unvented combustion devices \( S \) varies greatly depending on appliance type, primary aeration rate, fuel input rate, appliance use pattern, fuel type, etc. (Billick et al. 1984). In order to study some of the factors affecting source emissions, as well as to acquire a data base, two unvented combustion devices were chosen. One was a popular multiwick non-pressure kerosene stove (2.5 million users) and the second an LPG (liquefied petroleum gas) stove of a popular burner design (8 million users) both supposedly conforming to the specifications laid down by the Bureau of Indian Standards (I.S.2980 1979; I.S. 4246 1984).

The kerosene stove operates with a blue flame and gives high heat output, as in a gas stove. It has two concentric perforated cylinders between which the wicks emerge from the burner upon raising the wick lever. The annular space between the perforated cylinders serves to draw combustion air from either side for mixing with fuel vapors and for directing the flame upward.

EXPERIMENTAL DETAILS

The direct flue gas measurement technique (Yamanaka et al. 1979; Dave 1984) was adopted with an enclosed hood and duct assembly for capturing all the emissions, which were drawn out of the 100 mm diameter duct by means of a 100 mm diameter exhaust fan (Figure 1). The duct gas velocity was measured by a calibrated airflow meter sensor of 100 mm diameter, kept at right angles to the direction of duct gas flow (Figure 2). The typical duct airflows induced by the exhaust fan were of the order of 0.008 m\(^3\)s\(^{-1}\). The experimental set-up was enclosed by a specially erected chamber to prevent reentry of exhausted gases into the duct. The sampled gases were also conducted out of the chamber. To verify whether all the emissions were collected by the hood, in-chamber background levels of the same pollutants were monitored for unusually high values arising from spillover of combustion gases into the chamber. The background concentrations were deducted from duct gas concentrations in arriving at the emission factors.

All 28 samplings for the kerosene wick stove and 60 samplings for the LPG stove were obtained, both with and without a pan, at different burn rates. An aluminium pan that was 175 mm in diameter and 93 mm deep, weighing 280 g, and a predetermined thermal load of 1.5 L of water were used in samplings done with the pan in place. In the case of the kerosene stove, the emissions were studied at three different fuel levels, since the level of fuel in the tank was reported to affect the performance according to the operation manual of the stove. In the case of the LPG stove, two burners of different power ranges, as normally used in a dual-burner gas stove, were tested. The methods of analysis (Katz 1977) adopted for different pollutants are shown in Table 1.

Figure 1 Enclosed hood and gas stove in place. A-exhaust fan, B-duct, C-pan supports, D-burner, E-knob, F-grommets, G-rubber tubing to cylinder. All dimensions are in millimeters.

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The MSA detector tube method was chosen due to practical difficulties in obtaining other instruments, using the NDIR technique, in the desired time frame. The emission factor determination, using this method, is outlined in Appendix A. A sampling period of one hour was adopted for all parameters. The flow measurement was done using calibrated rotameters or a soap bubble meter for flow rates below 0.005 L·s⁻¹.

The fuel consumption rate was gravimetrically determined for the kerosene stove. In the case of the LPG stove, the burners were calibrated using a portable 5 kg LPG cylinder at three flame settings—minimum, medium, and maximum—in terms of grams of liquid-phase LPG consumed per hour at each setting. Only these settings were used in the study of emission factors. While it was not possible to replicate specific burn rates in the kerosene stove, it was comparatively easier in the gas stove. The standard deviation of the burn rates, at medium and maximum settings, is somewhat high as compared to the minimum setting for both burners (Table 2). This could be due to the absence of any clear-cut boundaries in the gas knob demarcating the medium and maximum flame conditions. In the “sim” setting, however, the simmer orifice controls the flow. The observed deviations here could be due to minor fluctuations in the flow of gas from the cylinder.

The burn rates were determined as described below:

The 5-kg cylinder was preweighed and connected to the stove by means of the pressure regulator. The regulator knob was opened with the stove knob still in the OFF position. A lighted match was brought near the burner and the stove knob was turned on. The desired stove setting was always approached from the maximum setting. After operating the stove for the desired duration with the stove knob still in the ON position, the regulator knob was turned off and the residual gas allowed to burn in the stove. The cylinder was then disconnected and weighed again. From the difference in weights of the cylinder, gas consumption per unit time was calculated.

In the case of samplings done with the pan in place, the sampling was started soon after blue flame adjustment was made and the pan was placed on the stove. For samplings with an open flame, sampling was started soon after the stove was lit. The objective, in the latter case, was to estimate the total emissions without the pan. The pollutants emitted during the short period of lighting and blue-flame establishment, as in the case of the kerosene stove, were not monitored separately since they were found to be below detectable limits of the methods chosen for analysis.

The average net calorific values of 42,810 kJ·kg⁻¹ of kerosene and 45,640 kJ·kg⁻¹ of liquid phase LPG were used in calculating the emission factors in terms of µg·kJ⁻¹. The sulfur content of the kerosene was determined as 0.07% by weight using the Raney Nickel method (IOM 121 1986) in a petroleum research facility. In fitting the emission pattern curves, linear, exponential, power, and logarithmic regres-

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**TABLE 1**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Method of Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nitrogen dioxide</td>
<td>Saltzman's colorimetric method</td>
</tr>
<tr>
<td>Nitric oxide</td>
<td>Saltzman's colorimetric method after oxidation to NO</td>
</tr>
<tr>
<td>Sulfur dioxide</td>
<td>West &amp; Gaeke's method</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>Chromotropic acid method</td>
</tr>
<tr>
<td>Carbon monoxide</td>
<td>MSA indicator tube method</td>
</tr>
<tr>
<td>Particulate matter</td>
<td>Gravimetric method using GF/C 47 mm filter</td>
</tr>
</tbody>
</table>

**TABLE 2**

<table>
<thead>
<tr>
<th>Burner</th>
<th>Setting</th>
<th>n</th>
<th>Burn rate of LPG Liquid g/h</th>
<th>Flame height cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Big</td>
<td>Sim</td>
<td>4</td>
<td>50.50 ± 0.24</td>
<td>2 – 3</td>
</tr>
<tr>
<td></td>
<td>Med</td>
<td>4</td>
<td>112.10 ± 1.85</td>
<td>8 – 10</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>4</td>
<td>239.02 ± 0.95</td>
<td>18 – 20</td>
</tr>
<tr>
<td>Small</td>
<td>Sim</td>
<td>4</td>
<td>47.11 ± 0.31</td>
<td>2 – 3</td>
</tr>
<tr>
<td></td>
<td>Med</td>
<td>4</td>
<td>93.12 ± 2.01</td>
<td>8 – 10</td>
</tr>
<tr>
<td></td>
<td>Max</td>
<td>4</td>
<td>158.07 ± 1.37</td>
<td>18 – 20</td>
</tr>
</tbody>
</table>

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**Figure 2** Main components of the airflow meter. A-press button switch, B-range selector, C-three-pin socket, D-indicator, E-three-pin plug, F-connecting cord, G-handle, H-magnet, I-angular vanes, J-bearings.

**Figure 3** Emission patterns for open flame operation of kerosene wick stove. Pollutants followed by Pearson's r in parentheses: ● - HCHO (0.79), ○ - NO₂ (0.73), △ - NO (0.80), Δ - CO (0.62), ■ - SO₂ (0.14), □ - PM (0.62).
sions were tested on a personal computer and the curve giving the best fit was chosen to represent the relationship between the two parameters.

RESULTS AND DISCUSSION

Referring to Figure 3, it can be seen that, with increasing burn rates, CO and particulate matter (PM) decreased, while all other pollutants showed an increase when the kerosene stove was operated without a pan. However, when the pan with a thermal load was used, the patterns changed, as shown in Figure 4. Here, except for HCHO and PM, all others showed an increase with increased burn rates. While the opposing trends of HCHO emissions in Figures 3 and 4 have something to do with the design of the combustion device, it may be seen that they are higher when the pan is used than when they were with an open flame operation. The scattergrams corresponding to Figure 4 are given in Appendix B (Figure B-1).

Nitric Oxide

The formation of NO is known to be dependent on the peak temperatures achieved during combustion. If the duct gas temperatures (DGT) are considered as surrogates for peak flame temperatures achieved in the open flame operation of the kerosene stove, then NO generation had been somewhat commensurate with the DGT, until 110°C (Figure 5). The fluctuating DGT could be due to a combined effect of fluctuating combustion efficiencies of the stove as well as airflows through the duct. The drop in NO at 120°C could have been due to the fuel mixture becoming rich and off-stoichiometric beyond the corresponding burn rate. This might be due to a design limitation in the appliance. The subsequent rise in NO value, at a DGT of 162°C, indicates that much of the NO in this case could have been formed in the post-combustion reactions (Edwards 1974).

In comparison, NO values appear to be the same, or somewhat more, with the pan than without it. It is possible that the pan bottom acts as a damper for air currents, thereby maintaining the reacting gases at a high temperature for a longer period, resulting in higher NO emissions. This does not happen in the LPG stove, perhaps because the burner design is different.

Figures 6 and 7 depict that, in the LPG stove, while NO formation in an open flame has a high correlation with burn rate, the correlation is poor when the stove is operated with a pan. This might be due to the prevalence of off-stoichiometric conditions at higher burn rates, owing to the burner design.

It can be seen from Table 3 that NO emissions from the LPG stove are greater than kerosene stove emissions by about 4 times when operated without the pan, and by 5 to 23 times when operated with the pan. Also, for the same range of burn rates, the LPG stove (small burner) produces 3 to 7 times

![Figure B-1 Scatter diagrams of overall emissions (µg·kJ⁻¹) against burn rates (kJ·min⁻¹) for covered flame operation of kerosene wick stove](image)

![Figure 4 Overall emission patterns for covered flame operation of kerosene wick stove (combined for all three reservoir fuel levels). Pollutants (Pearson's r): - HCHO (0.88), O - NO₂ (0.64), ▲ - NO (0.70), △ - CO (0.87), ■ - SO₂ (0.90), □ - PM (0.92).](image)

![Figure 5 Fluctuations in duct gas temperatures and nitric oxide emissions](image)
more NO without the pan, and 3 to 27 times more NO with the pan, than the kerosene stove.

Nitrogen Dioxide

When both oxygen and nitric oxide are present in the effluent gases, the nitric oxide will be oxidized to nitrogen dioxide as the gases cool. However, the rate is low and the residence time is much too short for the conversion to occur. NO₂ may also be formed due to flame quenching by a surface, e.g.,

\[ \text{NO} + \text{O} \xrightarrow{\text{Quench Surface}} \text{NO}_2 \]  

In either case, the NO₂ formed may be converted back to NO via the reaction

\[ \text{NO}_2 + \text{O} \rightarrow \text{NO} + \text{O}_2 \]  

which is also rapid at low temperatures (240–1800 K).

Thus, NO₂ exists only as a transient pollutant at flame
A comparison of Figures 3 and 4 reveals that through which combustion air diffuses. Reaction 2 may be dominating at the pan surface. The lower DGT, when the discused earlier. The correlation are more or less the same, both with and without the pan (Figures 7 and 6). The correlation between NO₂ and burn rate, in the former case, is poor (Pearson's $r = 0.62$), possibly due to the prevalence of the off-stoichiometric conditions discussed earlier. NO₂ emissions from the LPG stove are about 1 to 5 times greater without the pan, and about 2 to 3 times with the pan than those from the kerosene stove.

**Formaldehyde**

In the kerosene stove, formaldehyde formation may take place in a region where the pyrolytic precombustion products react with the hot air to form the final combustion products. At this stage, if the mixing of air and fuel vapor is not complete, the intermediate products could be transported away from the combustion zone, preventing further oxidation. Thus, HCHO emissions are higher when a pan is used than with open flame operation. The use of a pan could be inhibiting the mixture of combustion air and the pyrolytic precombustion products, especially at lower burn rates. The mixing may be better at higher burn rates, resulting in lower HCHO emissions.

Formation of HCHO in an LPG stove could be due to the inner layers of the flame not getting enough secondary air. The emissions were, however, 4 to 6 times less than kerosene stove emissions (Table 3).

**Particulate Matter**

Particulate matter (PM) in the kerosene stove is mostly a product of pyrolytic precombustion reactions and is emitted, as such, without further oxidation when mixing is not complete, as may occur at lower burn rates. The poor correlation observed in Figure 3 could be due to fluctuating mixing conditions in the concentric perforated cylinders. The emissions were more without the pan than with the pan. This could be due to the pan acting as a barrier to the free flow of particles in the buoyant effluent gas. PM emissions from the LPG stove were negligible and sporadic. No conclusions could be drawn from the observations available.

**Sulfur Dioxide**

Any fuel containing sulfur emits sulfur dioxide when burned. Generally its combustion is complete and hence stoichiometric concentrations of SO₂ are expected in the emissions. However, in the present study, the SO₂ emissions recovered were found to be proportional to the burn rate used despite the maintenance of the same sampling efficiency throughout, as shown in Figure 8. There was good correlation (Pearson's $r = 0.93$) between burn rates and SO₂ emissions both with and without a pan.

The literature was scanned to find out if similar observations were recorded in any of the studies done earlier. Indications to the effect that not all the sulfur burned was recovered as such were given by Woodring et al. (1985). They burned fuel containing 0.04% sulfur, but the maximum SO₂ recovery was about 8 µg·kJ⁻¹ as against the stoichiometric 17 µg·kJ⁻¹. No reasons were advanced for this phenomenon.

Since sulfur oxides and particulates are generally synergistic, it was decided to find out if any correlation existed between the two parameters. As shown in Figure 9, a strong negative correlation (Pearson's $r = 0.92$) existed between SO₂ and particulate matter, i.e., the higher the particulate matter, the lower the SO₂ and vice versa. This, coupled with the fact that activated carbon is used as an adsorbent to remove sulfur oxides from waste gases (Slack and Hollinden 1975), has led to the conjecture that some of the SO₂ formed could be getting adsorbed onto the particulate matter.

The stoichiometric SO₂ for a sulfur content of 0.07% in
Carbon Monoxide

In the present study, the burn rate was 110.91 kJ·min⁻¹, and the kerosene used had a latent heat capacity of 32.7 µg·kJ⁻¹. The maximum that could be recovered was 21.31 µg·kJ⁻¹ at a burn rate of 110.91 kJ·min⁻¹. It is presumed that the rest of the SO₂ could be getting adsorbed onto the particulate matter, in view of the strong negative correlation noticed between them. The fact that even atmospheric dust is known to adsorb SO₂ (Liberti et al. 1978; Tartarelli et al. 1978).

The range of SO₂ emissions from the kerosene stove is shown in Figure 3. The emissions were more or less the same for the respective burn rate ranges. In the open flame operation, burn rates correlated better with SO₂ emissions per minute (Figure 10) than with SO₂ emitted per kilo Joule (Figure 3).

No pattern could be discerned in SO₂ emissions from the LPG stove. The only sulfur source in LPG are the mercaptans added to give it a pungent odour for easy detection of the leaking gas. The quantities added are very small. Some of the heavier gases, like butane, may contain sulfur in disulfide form, but that supposition should be verified by detailed investigation to determine the species of sulfur compounds. As compared to the kerosene stove, SO₂ emissions from LPG combustion are negligible and of no concern.

Carbon Monoxide

Carbon monoxide can be produced either when the fuel vapor is not properly mixed or when the mixture is fuel rich. In this study, it has been observed that CO can also result when the flame temperature is rapidly lowered at a surface (flame quenching), thereby disrupting the oxidation reactions progressing in the flame. For this reason, CO emissions in open and covered flame operations cannot be compared using burn rate as a common criterion.

In open flame operation, CO generation takes place either due to improper mixing, as in the kerosene stove, or due to a lack of adequate primary air at low jet (sim) velocities, as in the gas stove. When a pan with a thermal load is used, the situation is different. Here, the rate at which the flame is quenched determines the rate of CO evolution. This was based on the observation that a good correlation (Pearson's $r = 0.88$) existed between CO emissions and the rate at which the latent heat of vaporization was lost to the atmosphere while boiling water, as shown in Figure 11.

Since the latent heat capacity of water is quite large (2270 kJ·kg⁻¹), it is postulated that the flame is quenched at the interface with the pan bottom because of the almost instantaneous transfer of a significant part of its heat into the duct, in the form of water vapor. The steep temperature gradient arising at the interface of the pan surface when water is boiling could be responsible for the rapid quenching of the flame. This was supported by the fact that CO evolution was sluggish until the water started evaporating.

Another supporting observation noted was that when all the water in the pan evaporated dry, the CO evolution also dropped drastically. Since there was no water to absorb and release this heat in vapor form, the heat was radiated into the duct, raising the duct gas temperature. Kane (1950) also observed that CO production increased with increasing impingement on the pan bottom. The reason attributed to that finding was increased interference with secondary air entrainment as the impingement area increased, resulting in less complete oxidation of CO to CO₂. While this may explain CO generation in the gas stove, it does not hold for the kerosene stove.

All these observations seem to justify the conclusion that, when a stove is operated with a pan in place and a thermal load of water, irrespective of how efficient the combustion may be, CO evolution does take place. It is a function of the rate at which the flame gets quenched at the interface with the pan bottom. This may, however, depend on the pan bottom, its shape, size, material, and contents; in short, anything that changes the lower surface temperature of the pan.

For instance, when the pan contents were changed from water to sand, there was very little CO generation, as compared to water. CO generation was more rapid when an enameled white tray (360 by 300 by 60 mm) instead of an aluminium pan was placed over the LPG stove. The CO generation was even more rapid when a hemispherical aluminium pan was used. These aspects need further study, involving the measurement of surface temperatures and gas film tem-

![Figure 10](image1.png)  
Correlation between sulfur dioxide emission rate and burn rate in open flame operation of kerosene stove (Pearson's $r = 0.88$)

![Figure 11](image2.png)  
Correlation between carbon monoxide emissions and heat loss due to vaporization (Pearson's $r = 0.88$)
CONCLUSIONS

Several points emerge from this work. The first observation is that both the kerosene and LPG stoves emit pollutants which are harmful to health. While the kerosene stove dominates in SO₂, HCHO, and PM emissions, the LPG stove dominates in NOₓ emissions. CO emissions are a common factor for both the stoves. Secondly, these emissions do fall into a pattern—increasing or decreasing, linear or curvilinear—in relation to burn rate and are affected by the use of a pan. Third, there could still be other factors that influence the emissions, as in the case of CO. Last, but not least, there appears to exist an optimum range of burn rates, especially in the case of the kerosene stove, as indicated by the shaded areas between which some pollutant emissions would be at an optimum (Figures 3 and 4). This optimum range is restricted to open flame operation of the big burner only in the case of the LPG stove (Figure 6).

It is to be cautioned that the emission patterns observed could be specific to the stoves and the pan used in this study. However, it throws open certain possibilities that might result in an abatement of emissions at the source. For instance:

a) The optimum range of burn rates observed from the emissions viewpoint could also exist in the unvented combustion devices (UCD) used in developed countries. This might help the manufacturers specify the optimum range of burn rates between which a UCD may be operated. Alternatively, for example, rate controllers may be installed on the gas manifolds for each burner, in the case of the gas stove. Whereas it is not possible to completely eliminate NOₓ emissions, it may be possible to reduce them substantially by this measure.

b) In the light of preliminary findings about CO generation, it might be possible to evolve an ideal utensil a la pressure cookers, pans, etc., for adoption on a mass scale, taking into account the size, shape, and material of the utensils vis-a-vis the emissions. In doing so, the cultural and aesthetic factors will, of course, have to be borne in mind.

c) The findings on CO evolution indicate that CO generation can be substantially reduced if the contents of the pan are covered, so as to reduce heat loss. This is significant because, in an assessment study done on the knowledge and management practices of urban housewives in regard to cooking fuels, it was found that a gap existed between knowledge and its application in fuel use (Ogale and Chaturvedi 1985). In several cases, the reluctance to light a match took precedence over switching off the burner of a gas stove in between operations. Then there are other undesirable habits like using smaller diameter vessels, an unjustified preference to always use the big burner, operating the stove at full flame even after the contents come to a boil, boiling without a lid, using excessive water for cooking, clogged burners, etc. Keeping the above factors in mind, it may be feasible to evolve health education strategies aimed at proper usage of the stoves both from pollution and thermal efficiency points of view as well as to avoid exposure to the pollutants.

Certain conclusions can be drawn from the present study:

1. Stray emission factors reported in published literature are difficult to use in characterizing the pollutant release rate of combustion devices. The emissions must be studied with respect to the operational factors influencing the use of the devices and the specific pollutant emission rates, as in the case of SO₂ and CO.

2. Both the kerosene stove and the LPG stove have a significant pollution potential in homes. Since there are no viable alternative "clean" fuels at present, it is necessary to take a second look at the design of these innocent-looking devices from the pollution angle and make modifications in the appliance design.

3. The present study also indicated the intimate complicity of combustion phenomena in the formation of pollutants in flames. Hence any source emission modeling effort must incorporate a synthesis of pollutant chemistry and combustion science principles.

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REFERENCES


APPENDIX A
METHOD OF CO DETERMINATION USING MSA DETECTOR TUBES

The indicator tubes, when fitted to the squeeze bulb and aspirated, were expected to show stains of different intensities corresponding to different volumetric percents of CO, depending on the number of squeezes; they were thus calibrated at a particular flow rate. In this case it was 0.1 liter per minute. Since the duct gas CO concentrations were not high enough to produce a stain in the tube in one or even five squeezes of the bulb (the maximum indicated on the color chart), it was decided to aspirate the gas through the tube at the same rate, 0.1 lpm, for an extended period of time until the stain showing 0.1% was achieved. This was done by connecting the CO indicator tube to the sampling system and plugging it directly into the G.I. duct. A thermocole (expanded polystyrene) sleeve was fitted to the tip of the tube going into the G.I. duct to prevent heating of the tube by hot gases. In each case, the time taken to reach 0.1% concentration stain was noted. This ranged from 5 to 60 minutes. Intermediate stain intensities were approximated to the nearest % CO concentration reading, or an average value was taken. All parameters measured at elevated temperature and pressure were reduced to their respective values at 25°C and 760 mm Hg. The concentration of CO in the duct was then calculated.

As an example:

Assume that a 0.1% concentration was recorded in 10 minutes, at a flow rate of 0.1 liter per minute. The quantity of air sampled in 10 minutes is, therefore, 1 L. The standard volume of the sample is calculated to be 0.9 L at 25°C and 760 mm Hg. Therefore, the 0.1% concentration which should have been achieved in one squeeze (or 50 ml) of the bulb has been achieved after aspirating 900 ml. The CO concentration in the duct air is given by:

\[
\frac{50}{900} \times 0.1\% \times 10,350 \text{ ppm} \times 1145 \mu g \cdot m^{-3} = 65,837 \mu g \cdot m^{-3}
\]

where

\[
10,350 \text{ ppm} = 1\% \text{ CO}
\]

\[
1145 \mu g \cdot m^{-3} = 1 \text{ ppm CO}
\]

Knowing the duct gas rate of flow (= duct gas velocity \(\times\) cross-sectional area of the duct), the mass of CO emitted can be calculated. This, when divided by the fuel consumed in unit time, yields the emission factor.