Emission of Particulate Matter While Applying the Envirox™ Additive

FRANTISEK BOZEK, JAROMIR MARES, MILOS BOZEK
Civil Protection Department
University of Defence
65 Kounicova, 662 10 Brno
CZECH REPUBLIC
frantisek.bozek@unob.cz http://www.vojenskaskola.cz

JIRI HUZLIK
Transport Research Centre
2557/33a Lisenska, 636 00 Brno-Lisen
CZECH REPUBLIC
jiri.huzlik@cdv.cz http://www.cdv.cz

Abstract: - The values of emissions of particulate matter (PM) were monitored in the older type of Diesel engine operating on a testing bench and then compared with the data acquired under similar conditions when the Envirox™ additive was applied. It was found out that the additive based on cerium dioxide nanoparticles reduces emission of PM. The dependence of PM emission on reduced torque $\tau_{\text{R}}$, engine power $P$ and revolutions $r$ was observed as well.

Key-Words: - Additive, air, cerium dioxide, Diesel fuel, emission factor, nanoparticle, particulate matter, pollutant.

1 Introduction

Escalating activities of human society aimed at ensuring a higher standard of living bring a number of negative externalities. Among the important and by the lay and professional community strongly discussed externalities belongs the increase of burden to the atmosphere caused by industrial and agricultural activities, energy production, waste management and household management. Mobile assets, including primarily transport of passengers and cargo [1, 2], significantly contribute to the increase in ambient air pollution as well.

A number of other hazardous substances are produced [2] apart from the quantitatively dominant pollutants, which include carbon dioxide (CO$_2$), carbon monoxide (CO), mix of nitrogen oxides (NO$_x$), volatile organic compounds (VOCs) and particulate matter (PM). Emitted pollutants often have non-quantifiable impacts on morbidity and mortality of the population, ecosystem function and value of social assets [3].

As transportation is currently one of the world's most dynamically developing sectors, it is necessary to pay enormous attention to the reduction of emissions. This requirement is raised by the use of cars at the expense of public transport and permanently increasing ratio of road haulage transport to rail transport [1].

The paper is focused on evaluating the quantities of PM emissions while applying conventional Diesel fuels without and with the Envirox™ additive.

2 The Analysis of Current State

Recently, considerable attention is devoted to the health effects of emissions of suspended PM. The danger lies mainly in its ability to bind a number of toxic inorganic and organic pollutants to the surface. The biggest problem is represented by particles smaller than 2.5 µm which cannot be captured in the upper respiratory tract. PM$_{2.5}$ and smaller factions therefore penetrate into the alveoli and as the efficiency of their removal is limited, inhalation causes serious health problems [1].

Epidemiological studies have attributed not only short but also chronic effects to PM, especially in the long term exposure, even at lower concentrations. Short-term effects are reflected in the form of mechanical damage to the cornea, difficult breathing, deterioration of health among asthmatic patients, and lung tissue damage leading to the fibrillation of lungs [1, 4]. Long-term effects can be seen in the increased incidence of bronchitis, cardiovascular and reproduction disorders and, in case of extreme exposure, cancer of mainly respiratory organs [3, 5].

The amount of pollutants, including PM, emitted by a truck is dependent on many factors. If driving style, terrain and weather conditions are not considered, and then the quantity of produced emissions depends mainly on the following factors [2]:

a) The type of engine and its technical parameters;
b) Principles of oxidation catalyst;
c) The type and amount of biodiesel added to fuel;
d) The composition and quality of Diesel oil;
c) The type and composition of additives added to the basic fuel.

The calculation of the \( i \)-th pollutant emission is based on the knowledge of an emission factor \( E_{i m} \) [g kg\(^{-1}\)] which is given by formula (1) as the weight of the \( i \)-th pollutant per a unit mass of consumed fuel \( F \) [2, 6]:

\[
E_{i m} = m_i \times m_F^{-1} = y_i^d \times \frac{M_i \times n_i^d}{M_F \times N_F}
\]

where \( m_i \) [g] is the mass of the \( i \)-th pollutant, \( m_F \) [kg] weight of fuel, \( M_i \) molar molecular weight of the \( i \)-th pollutant [g mol\(^{-1}\)], \( M_F \) [kg mol\(^{-1}\)] molar molecular weight of fuel, \( n_i^d \) [mol] or \( N_F \) [mol] substance amount of dry exhaust gas or consumed fuel, and finally \( y_i^d \) molar fraction of the \( i \)-th pollutant in dry exhaust gases.

The industry producing and supplying fuels operates with a number of additives that can be added to the Diesel fuel. These can be divided into three main areas according to their nature [7]:

a) Refinery additives;

b) Safety increasing and legally required additives;

c) Additives for the improvement of technical parameters and increase of fuel performance.

The Envirox™ is a dispersion of CeO\(_2\) nanoparticles in aliphatic and cycloaliphatic hydrocarbons. This Diesel fuel additive should, according to promotional materials of Oxonica (now Energenics) company, improve more than only the technical parameters of fuel [7]. Unlike conventional additives, Envirox™ is advertising efficiency throughout the combustion process, because most of the common ingredients decompose under the thermodynamic conditions prevailing in the engine combustion chamber. The Envirox™ enables engine to gain more energy from fuel and therefore reduce its consumption, remove the residual soot deposits in engine combustion chamber while minimizing the formation of some contaminants [8]. Chemistry of combustion process is presented in equation (2) for the more efficient use of fuel energy, in equation (3) for the elimination of carbon deposits in engine combustion chamber and equations (3-6) for the reduction of pollutants in emissions [2].

\[
\begin{align*}
(4x+y) \text{CeO}_2 + 2x \text{CO}_2 + y \text{H}_2\text{O} \\
4 \text{CeO}_2 + C &= 2 \text{Ce}_2\text{O}_3 + \text{CO}_2 \quad (3) \\
2\text{Ce}_2\text{O}_3 + \text{CO} &= \text{Ce}_2\text{O}_4 + \text{CO}_2 \quad (4) \\
2 \text{Ce}_2\text{O}_3 + 2 \text{NO} &= 4 \text{CeO}_2 + \text{N}_2 \quad (5) \\
4 \text{Ce}_2\text{O}_3 + 2 \text{NO}_2 &= 8 \text{CeO}_2 + \text{N}_2 \quad (6)
\end{align*}
\]

\[
\text{CeO}_2 \text{ regeneration catalyst is carried out in accordance with chemical formula (7).}
\]

\[
2 \text{Ce}_2\text{O}_3 + \text{O}_2 = 4 \text{CeO}_2
\]

Statistically validated operational tests carried out by Oxonica provide evidence that the recommended dosage of 5 to 10 ppm w/w CeO\(_2\) can achieve relevant reductions in fuel consumption (about 5-12\%) while reducing the emissions of CO\(_2\), CO, NO\(_x\), C\(_6\)H\(_4\) and particulate matter. The additive is also compatible with all common Diesel additives [8, 9].

### 3 Problem Solution

#### 3.1 Applied Methods and Devices

Tests to determine emission levels were carried out on the VOP-026 Sternberk company engine testing bench with the Schenck 0-900 kW electric eddy current brake operating in the range of 0-6.0×10\(^3\) revolutions min\(^{-1}\). NM-54 Diesel was used as the primary fuel, which was mixed with 2.5×10\(^4\) volumes of additives in an alternative version of comparative tests. The concentrations of CeO\(_2\) found in Diesel fuel by inductively coupled plasma atomic emission spectroscopy was 7.6 ppm w/w, which corresponds to Envirox™ suppliers’ requirement.

Diesel engine with the following characteristics was used for the test: Tatra T3 930-31 four stroke, naturally-aspirated engine with direct injection, air-cooled, engine cylinder capacity of 1.9×10\(^4\) cm\(^3\), cylinder diameter/stroke 120/140 mm, OHV distribution and a compression ratio of 1:16. The engine had 12 cylinders in two separate lines at 90°. Rated engine output was 235 k\(W\) ± 10\% at 2.2×10\(^3\) min\(^{-1}\) with a maximum torque of 1.13×10\(^3\) N m at revolutions 1.4×10\(^3\) ± 200 min\(^{-1}\).

It was necessary to know the concentration of combustion gas emissions in order to calculate the \( E_{i m} \) emission factor for the PM. The gas emission testing was performed by a combined device called ECOM-JN for analyzing the combustion gas composition, equipped with electrochemical sensor of the English company City Technology, which enabled the determination of CO, NO, NO\(_2\) and O\(_2\) concentration. Ranges and uncertainty in the determination of individual quantities are listed in Table 1.

<table>
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<tr>
<th>Pollutant</th>
<th>Range [ppm]</th>
<th>Uncertainty of measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO</td>
<td>0-2.0×10(^3)</td>
<td>2 %</td>
</tr>
<tr>
<td>NO(_2)</td>
<td>0-2.0×10(^2)</td>
<td>2 %</td>
</tr>
<tr>
<td>CO</td>
<td>0-1.0×10(^3)</td>
<td>2 %</td>
</tr>
<tr>
<td>O(_2)</td>
<td>0-2.1×10(^3)</td>
<td>2 %</td>
</tr>
</tbody>
</table>

Sample of combustion gas was taken by a vacuum pump tube probe analyzer. The current air mass was led
from the probe tube by unheated tube to filters and water separators analyzer and then to pollutant sensors. It was possible to determine the concentration of CO and NO, by applying unheated tube between the probe and analyzer as possible combustion gas condensation on the way did not affect their values.

C₃H₄ content was tested by analyzer operating on the principle of flame ionization (FID). The principle is based on the effect that the C-H bond gets ionized during the burning of hydrocarbons in the hydrogen flame of the analyzer burner combustion chamber. If the electrodes placed in the burner are energized, the value of flowing current is proportional to the number of free ions including organic matter content in the sample. The sample of gases was transported into the FID unit through a heated tube of analyzer vacuum pump.

The manual gravimetric method was used to measure the concentration of PM. The method consists in inserting the probe into the measuring point and extracting the dusty air mass sample, so that there was the same rate of bearing air mass (so-called isokinetic sampling) in its aperture and around the probe. Extracted and measured volume of sampled air mass has been deprived of solid additives on a special filter and later, in the prescribed manner, the weights of the captured particles were determined. An extraction set with zero probe was used for the measurement, with a filter assembly positioned outside the pipe. Setting the isokinetic sampling with zero probe was achieved by regulating of exhaust gas quantity so that the pressure differential at zero probe reached zero. The planar filter used by the Amersil-filpap of Steti company is manufactured from 100 % borosilicate glass microfibers without organic bonding agents. The paper is resistant to temperatures up to 770 K and is able to retain greater amount of impurities before any increase in resistance to the passage of filtered media.

Measurement of ventilation parameters was carried out according to standard operating procedure [10]. The “L” shaped Prandtl probe of 8 mm diameter was used to measure the rate of combustion gas. The probe was fixed in the middle section of the measuring extension.

The location of measuring point, where the combustion gas velocity was measured with the Prandtl probe simultaneously with gas temperature measured with K (NiCr-Ni) thermocouple, results from Figure 1.

![Fig. 1 Location of sampling probes](image)

3.2 Outcomes and Discussion

3.2.1 Mass balance

The calculation of PM emission factor was based on relation (8) characterizing the fuel combustion, mass balance of carbon, hydrogen, nitrogen, and oxygen, the amount of dry gases substance entering and exiting the combustion process and on the measured concentration of O₂ and CO, NO and NO₂ pollutants. Fuel combustion in the engine was considered under simplified conditions in the absence of trace amounts of polycyclic aromatic hydrocarbons, N₂O, NH₃, SO₂ etc. Mass balance was described in detail in our paper focused on determination of emission factors for gaseous emissions [2].

$$ CO₂ + a×CO + (b/2) H₂O + d×(1-w) NO₂ + e×w NO \quad (8) $$

amount of i-th component in wet output, $N_v^d$ [mol] is the substance amount of dry gases entering into the combustion process and $n_i^d$ [mol] is the substance amount of dry gases exiting the process.

3.2.2 Molar fraction of unmonitored components

Because the concentration of water vapor and CO₂ in combustion gases were not monitored it was necessary to express their molar fractions from the mass balance. Equations (12) and (13) were obtained for the molar fraction $y_{H, O}^d$ and $y_{CO₂}^d$ respectively, as it was presented earlier in our WESSEX paper as well [2].
Concentration of water vapor from equation (18) can be expressed by formula (19):

\[
y^d_{H,O} = \beta \times \frac{Y^d_{O_2} - y^d_{O_2} + (1 - Y^d_{O_2}) \times \frac{y^d_{CO}}{2} - \left(1 - \frac{Y^d_{O_2}}{2}\right) \times y^d_{NO} - \frac{y^d_{NO}}{2} - Y^d_{O_2} \times y^d_F}{1 + \omega \times (1 - Y^d_{O_2})}
\]  

(12)

\[
y^d_{CO_2} = \frac{Y^d_{O_2} - y^d_{O_2} - \left[\omega \times (1 - Y^d_{O_2}) + \frac{1 + Y^d_{O_2}}{2}\right] \times y^d_{CO} - \left(1 - \frac{Y^d_{O_2}}{2}\right) \times y^d_{NO} - \frac{y^d_{NO}}{2} - Y^d_{O_2} \times y^d_F}{1 + \omega \times (1 - Y^d_{O_2})}
\]  

(13)

3.2.3 Conversion of concentrations on dry air

The formula (14) was used to convert the concentration of i-th component of the wet gas emissions on dry gas, where \(n\) [mol] is the substance amount of wet exhausted gases and other symbols have the same meaning as above.

\[
y^d = y_i \times n \times (n^i)^{-1}
\]  

(14)

Relation (15) results from the mass balance:

\[
n = n^d + N_{H,O} + N_{O_2} = 1 + \frac{N_{H,O}}{n^d} + n^d
\]  

(15)

\[
\delta = -\frac{n^d}{n} + 1 + \frac{\beta}{2} \times \left[ B \times \left(\frac{y^d_{CO}}{2} - \frac{y^d_{NO}}{2} + y^d_F - 1\right) + X \times \left(1 - \frac{y^d_{O_2}}{2} - \frac{y^d_{NO}}{2} - y^d_F\right)\right] \times (\omega + B + X)^{-1}
\]  

(18)

3.2.4 Calculations of water vapor concentrations

Concentration of water vapor \(Y^d_{H,O}\) converted into dry air, which is needed to calculate the conversion factor \(\delta\) from equation (18) can be expressed by formula (19):

\[
Y^d_{H,O} = x^d_{H,O} \times M_{H,O}^d = \frac{\varphi \times p_{SAT}^{H,O}}{p_{STC} - \varphi \times p_{SAT}^{H,O}}
\]  

(19)

where the symbol \(x^d_{H,O}\) represents the specific humidity of the air, \(M_{H,O}^d\) [g mol\(^{-1}\)] molar molecular weight of dry air, \(M_{H,O}^d\) [g mol\(^{-1}\)] molar molecular weight of water, \(\varphi\) [%] relative humidity of the air, \(p_{SAT}^{H,O}\) [Pa] the partial pressure of saturated water vapor at temperature \(T\) [K] and finally \(p_{STC} = 1.013 \times 10^5\) [Pa] pressure at standard conditions. In accordance with the literature [11] it is possible to calculate the partial pressure of saturated water vapor in the temperature interval from 273 to 473 K according to equation (20), where symbols \(C_8, C_9, C_{10}, C_{11}\), and \(C_{12}\) mean tabulated constants for air temperature \(T\) [K].

\[
\ln p_{SAT}^{H,O} = C_8 \times T^{-1} + C_9 + C_{10} \times T + C_{11} \times T^2 + C_{12} \times T^3 + C_{13} \times \ln T
\]  

(20)

The formula (24) for constant \(\gamma\) was derived using equations (21) and (22) and the equation (25) for constant \(\omega\) after substituting \(\gamma\) into relation (9):

\[
\gamma = \frac{\psi_H \times A_C}{\psi_C \times A_H} \quad (21)
\]

\[
\gamma = \frac{\psi_O \times A_C}{\psi_C \times A_O}
\]  

(22)

\[
\mu_F = A_C + \beta \times A_H + \gamma \times A_O = 100 \times A_C \times \psi_C^{-1}
\]  

(23)

where \(\psi_C, \psi_H, \psi_O\) means a relative content of carbon, hydrogen and oxygen in the fuel, \(A_C, A_H, A_O\) [g mol\(^{-1}\)] corresponding molar atomic weights and constants \(\beta\) and \(\gamma\) have the same meaning as in equation (9).

The molecular weight of fuel \(\mu_F\) related to one carbon atom is given by equation (23):

\[
\mu_F = A_C + \beta \times A_H + \gamma \times A_O = 100 \times A_C \times \psi_C^{-1}
\]  

(23)
3.2.6 Calculation of emission factors

Equation (27) is valid for the emission factor \( E_{f, PM} \) [g h\(^{-1}\)] of particulate matter in mass units related to time unit \( t \) [h]:

\[
E_{f, PM} = \frac{m_{PM}^t}{t} = \frac{m_{PM}^t}{V_{STC}^t \times t} = \frac{C_{PM}^{STC} \times V_{STC}^{STC}}{V_{STC}^t \times t}
\]  

(27)

where \( m_{PM}^t \) [g] represents the mass of captured PM in time \( t \), \( V_{STC}^t \) [m\(^3\) h\(^{-1}\)] means the volume of exhaust gases under standard conditions released during time \( t \), and finally \( C_{PM}^{STC} \) [g m\(^{-3}\)] the concentration of PM in exiting gas at standard conditions (STC).

\[
V_{STC}^t = \frac{m_{PM}^t \times V^*}{y_{CO_2}^d + y_{CO}^d + y_{F}^d} + \frac{1}{\mu_F \times t} + \beta \times \frac{B \times (y_{CO}^d - y_{NO_2}^d + 2y_{F}^d - 2) + X \times (2 - 2y_{O_2}^d - y_{NO_2}^d - y_{NO}^d) - 2y_{F}^d}{4 \times (\mu_F \times t) \times (X + \omega \times B)}
\]

(28)

Substituting for \( V_{STC}^t \) in the equation (27) it can be calculated \( E_{f, PM} \). The relation (30) follows from equation (1), from which it is possible with the known value of \( E_{f, PM} \) and the measured fuel consumption \( m_f^t \) per time unit \( t \) to express an emission factor \( E_{f, PM} \) related to the unit of consumed fuel mass.

\[
E_{f, PM} = \frac{m_{PM}^t}{m_f^t} = \frac{m_{PM}^t}{t} \times \frac{t}{m_f^t} = E_{f, PM} \times \left( \frac{m_f^t}{t} \right)^{-1}
\]  

(30)

Calculated values of \( E_{f, PM}^m \) under varying conditions.

It is evident that it will be necessary to estimate \( V_{f, STC} \) for calculating the emission factor \( E_{f, PM} \). The relation (28) below is valid for \( m_f^t \times t^{-1} \), which represents the amount of consumed fuel per time \( t \), while accepting the mass balance for consumed fuel and the equation for molar volume of ideal gas \( V^* \) [mol] under standard conditions.

\[
\frac{m_f^t}{t} = \frac{M_f \times \left( \frac{y_{CO}^d + y_{CO}^d + y_{F}^d}{a} \right) \times V_{STC}^{STC}}{V^*}
\]

(28)

Assuming the validity of equations (18) and (26), the formula (29) for \( V_{f, STC} \) follows from relation (28):

\[
V_{f, STC} = \frac{m_f^t}{t} \times (X + \omega \times B)
\]

(29)

of engine operation in the use of Diesel NM-54 with or without additive Envirox™ are presented in Table 2 together with the measured concentrations of pollutants and the concentration of O\(_2\). From there it is obvious that the PM emissions decreased by about 17% when the additive was applied to the diesel fuel. Such a decrease even slightly exceeds the value interval stated by the Oxonica company. The above mentioned company carried out long-term tests in urban and suburban transport and it advertises that the Envirox™ additive reduces PM emissions from 9 to 16% [9] depending on different operating conditions and the type of engine.

### Table 2: Engine parameters, concentration of pollutants and PM emission factor with and without Envirox™ additive

<table>
<thead>
<tr>
<th>Test</th>
<th>ML [%]</th>
<th>( r ) [min(^{-1})]</th>
<th>( TM_R ) [N m]</th>
<th>( T_{CG} ) [K]</th>
<th>( T_{EP} ) [K]</th>
<th>( P ) [kW]</th>
<th>( FC ) [%]</th>
<th>( O_2 ) [ppm]</th>
<th>( CO ) [ppm]</th>
<th>( NO_x ) [ppm]</th>
<th>( C_H_4 ) [mg m(^{-3})]</th>
<th>( PM ) [mg m(^{-3})]</th>
<th>( \pm_{fPM}^m ) [g kg(^{-1})]</th>
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<tbody>
<tr>
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<td>2201</td>
<td>934.6</td>
<td>772</td>
<td>832</td>
<td>215.4</td>
<td>50.2</td>
<td>7.9</td>
<td>2165.1</td>
<td>2049.9</td>
<td>25.1</td>
<td>11.0</td>
</tr>
</tbody>
</table>

**Note:** Table 2 shows the calculated values of \( E_{f, PM}^m \) under varying conditions.
The maximum fuel savings of approx. 1.5% were observed when the additive was applied and \( TM_R = 620 \text{ N m}, P = 120 \text{ kW} \) at \( r = 1800 \text{ min}^{-1} \). This fact is in conflict with the presented data by Oxonica, which, depending on engine type and operating conditions, declares reduction in fuel consumption from 5 to 12%.

At the same time the functions of the PM emission factor on the reduced torque \( TM_R \), revolutions \( r \) and engine power \( P \) was observed. Dependencies show a minimum PM pollution in the interval of \( TM_R \in (600; 800), \text{ N m} \) at the revolutions \( r = 1800 \text{ min}^{-1} \) and at engine power \( P \in (130; 180) \text{ kW} \) which is in compliance with theoretical expectations. The example in Figure 2 illustrates the graphical dependence of emission factor \( E_{f_{\text{PM}}} \) as a function of engine power \( P \). Format of the trend line was evaluated by linear regression through the polynomial of second degree with the corresponding regression equation and reliability coefficient \( R \).

4 Conclusion

The methodology of measuring and calculation of PM emissions in engine exhaust was developed. It was discovered that the Envirox™ additive based on dispersed CeO\(_2\) nanoparticles reduces the value of the PM emission by approx. 17%. Nevertheless, the consumption reduced by 5-12% as declared by the Oxonica company was not verified. The maximum fuel savings of 1.5% was found out only under optimum conditions of engine operation.

At the same time it was monitored there is dependence of emission factors on reduced torque, engine power and engine revolutions. Except for the increase in PM emission at revolutions \( r = 1800 \text{ min}^{-1} \) the mentioned functions were in accordance with theoretical expectations.

Disagreements with the data reported by Oxonica on minimization of fuel consumption when applying the additives and the dependence of emission factor \( E_{f_{\text{PM}}} \) on engine revolutions when operating the diesel fuel without an additive will have to be verified in further tests on a new type of engine and after sufficiently long period of engine operation with the efficient Envirox™ additive.

References
