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RESEARCH ARTICLE

Experimental Investigation of Discoloration Generated by a CI ICE's Exhaust Gas on Various Stone Types

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Abstract

The effect of particulate matter - as a component of an internal combustion engine's exhaust gas - on 8 different types of construction materials have been studied under laboratory conditions. Our aim was double. On the one hand it was to investigate the degree of discolouration, whether there is a difference between the 8 types of rock in the same exposure, after laboratory contamination. On the other hand to measure the mass effect of particulate on the rocks. For testing the adverse effects of diesel soot and particulate matter on stones a small scale laboratory exposure chamber was constructed and built in the exhaust system of the engine. A compression ignition engine was used to pollute directly the set of stones. Mass and colour measurement tests have been carried out on the stones before and after the exposure. The result is as expected small difference in the mass properties and a greater changes in terms of colour measurement.

Keywords

particulate matter, construction materials, CIELAB coordinates, compression ignition engine, weight loss

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1 Introduction

Natural stones are frequently used outside and inside. One of the key problems of external use is the discoloration of building facades. The colour changes can be attributed to air pollution, in the form of deposition of pollutants which lead to blackening of building stone (Bonazza et al., 2005; Grossi and Brimblecombe, 2007; Urosevic et al., 2012). The blackening process clearly related to particulate matter (Amoroso and Fassina, 1983). The blackening and formation of soiling layer have been described for various stone types including marble (Moropoulou et al., 1998; Pozo-Antonio et al., 2017), limestone (Fobe et al., 1995; Maravelaki-Kalaitzaki and Biscontin, 1999; Amoroso and Fassina 1983), travertine (Török, 2008) and even on volcanic rocks (Germinario et al., 2017; Graue et al., 2013). The soiling is related to particle deposition and incorporation into the newly formed gypsum layers (Sabbioni, 1995). The colour change reflects air quality and thus millennium long changes can be also recorded (Brimblecombe and Grossi, 2009). In the past decades vehicles were the main causes of urban stone decay, however air pollution can cause damage in stone structures located in rural settings (Török et al., 2011; Graue et al., 2013). The colour change and sulfation process can be modelled under laboratory conditions where carbonate samples are exposed to exhaust gas (particulates) and SO_x (Rodriguez-Navarro and Sebastian, 1996). The changes can be recorded by testing newly formed mineral phases and also by measuring colour changes (Grossi and Brimblecombe 2007). Spectroscopy can be used for studying these variations in colour of stones. It is also a useful tool to measure the differences by before-after tests and as well as the grade of weathering (Nagano and Nakashima, 1989).

Our basic aim was to investigate the aesthetic effect of air pollution derived from road vehicle's engine on stones widely used for construction and decoration. The main question is how this induced pollution contribute to accelerated weathering and how the exhaust gas related particulate matters influence the colour of stone slabs. Similar chamber test was applied to evaluate the role of particulate matter coming from vehicular sources on stone sulfation (Rodriguez-Navarro and Sebastian, 1996). This paper focuses on colour change rather than on mineralogical alterations of stone slabs having different composition.

2 Materials and methods

2.1 Stones

The tests were carried out on 8 different stone types. These types of stones have been chosen because they are used widely and they have physical and chemical properties which are very different. These 8 types of natural stones are used in the largest quantity for constructions and decorations in Hungary (Török, 2007; Siegesmund and Török, 2011). Except for Carrara marble from Italy and Mauthausen granite from Austria, the other 6 stone types were obtained from Hungarian quarries. All the specimens have cylindrical shapes with a diameter of 3 cm. The tested stone samples can be seen on Fig. 1.

The selected types of stones (and their places of origin) are as follows (from left to right on Fig. 1):

- rhyolite tuff (Sirok)
- porous limestone (Sóskút)
- andesite (Gyöngyös)
- granite (Mauthausen)
- marble (Carrara)
- sandstone (Romhány)
- travertine (Süttő)
- non-porous limestone (Tardos).

2.2 Pollution of stones

A block of 8 different stone types was placed in a chamber, which was built in the exhaust gas system of a compression ignition internal combustion engine, so the exhaust gas of the engine was led through the chamber. In this case the exhaust stream could contact directly to the surface of the material specimens. The location of the exposure chamber can be observed in the Fig. 2, and at the same time the chamber with the stones is shown in the Fig. 3.

A compression ignition engine was used to generate exhaust gas to pollute directly the stones. The most important data of the engine are listed in Table 1. This engine was running on conventional diesel, which was purchased from a petrol station in Budapest.

Table 1 The most important parameters of the engine used for pollution

Engine manufacturer	RÁBA
Engine type	D10 UTSLL 190
Emission approval	EURO II
Fuel	diesel (MSZ EN 590)
Engine operating point	1300 rpm ; 50% load

The engine was operated for 10 hours (twice 5 hours) at one operating point. Operating parameters of the engine were controlled and the composition of the exhaust gas (gas phase components), smoke values and temperature of the exhaust

Fig. 1 Stone types before exhaust gas pollution

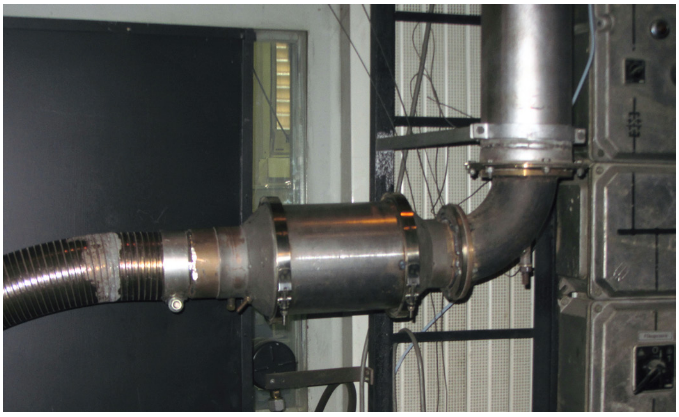

Fig. 2 Exposure chamber

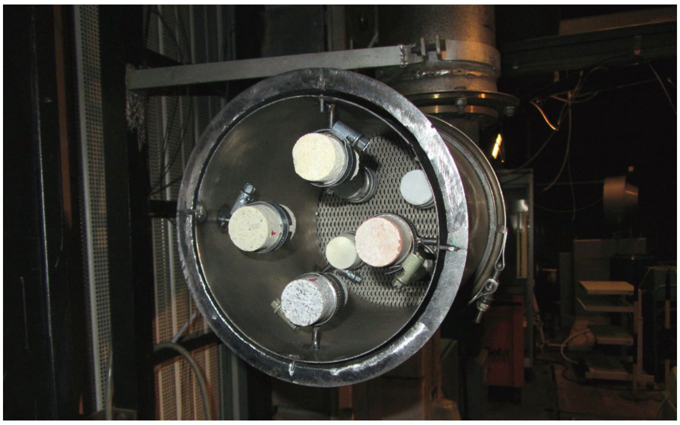

Fig. 3 Tested stone specimens in the chamber

gas were continuously measured during the test. The engine operating point was chosen due to the higher k value (Szabados and Bereczky, 2015; Szabados and Merétei, 2015; Ajtai et al., 2016).

The most important data of the engine operation can be read on the Table 2.

Table 2 Main exhaust parameters of the engine

Fuel consumption	3.91 g/s
Specific fuel consumption	225.55 g/kWh
Temperature of exhaust gas	330°C
Temperature in the chamber	260°C
Pressure in the chamber	1 – 2.5 Hgmm
Mass flow of exhaust gas	485 kg/h

The gas composition and the smoke values are listen in Table 3.

Table 3 Values of gas phase components and particulate relevant emission

CO	CO_2	THC	NO_x
[ppm]	[%]	[ppm]	[ppm]
146.5	6.02	32.8	742.2
NO	**O_2**	**FSN**	**k**
[ppm]	[%]	[mg/m^3]	[m^{-1}]
620.5	12.84	9	0.086

2.3 Test methods of polluted stones

The first parameter which has been determined on the polluted stones is colorimetric value (Choudhury, 2014; Antal et al. 2017). These values were recorded with a Konica Minolta Spectrophotometer CM-2600d. The specifications of the spectrophotometer are the following:

- Wavelength range: 360 nm to 740 nm (pitch: 10 nm)
- Measurement area: ø 8mm
- Observer: 2
- Illuminant: D65.

Measurements regarding the mass of the stones before and after the pollution have been carried out with the help of the analytical balance (Kern AES Analytical Balance).

3 Results

After the pollution, it can be seen by a visual inspection that each type of rock is blackened (as shown by Fig. 4).

A soot/particulate layer was formed on the external surface of the stones.

The blackening would be the most strongly on the face of the stones which are directly in the direction of flow. But it can be observed a deposition on the rear ends of stones due to different flow phenomena, mixing in the chamber. In case of stones having greater pores the soot congested the pores which are associated with the surface while particulates deposited on the surface as for solid stone probes.

The CIE (Commission Internationale de l'Eclairage) 1931 colour spaces are the first defined quantitative links between physical pure colours/wavelengths in the electromagnetic visible spectrum and physiological perceived colours in human colour vision. The chromaticity of a colour was then specified by the two derived parameters of x and y (Fig. 5) (Hunt and Pointer, 2011).

The CIELAB colour space is a colour-opponent space with dimension L* for lightness and a* and b* for the colour-opponent dimensions (a: redness-greeness, b: yellowness-blueness), based on nonlinearly compressed (CIE xyz colour

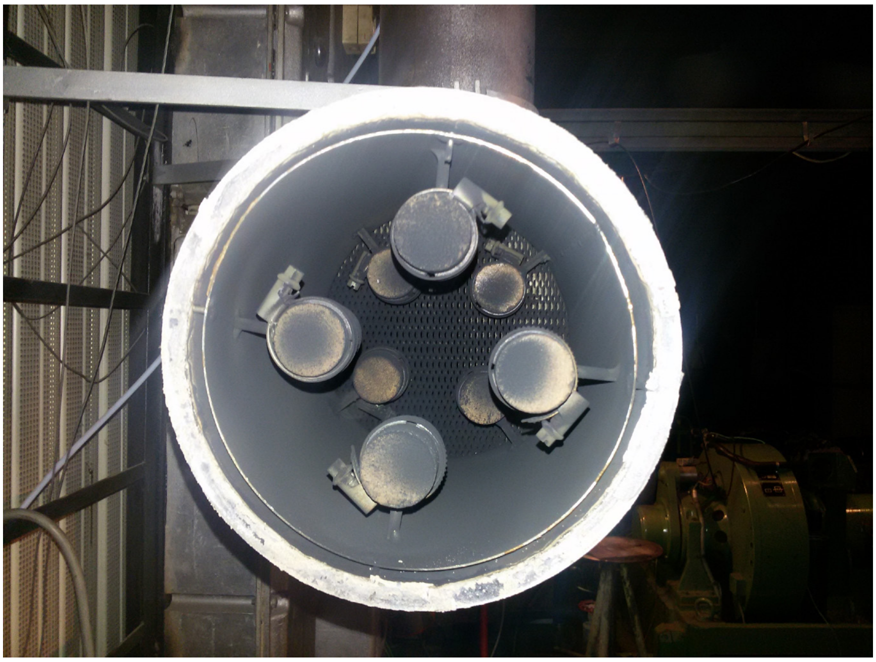

Fig. 4 Tested stone specimens after the pollution

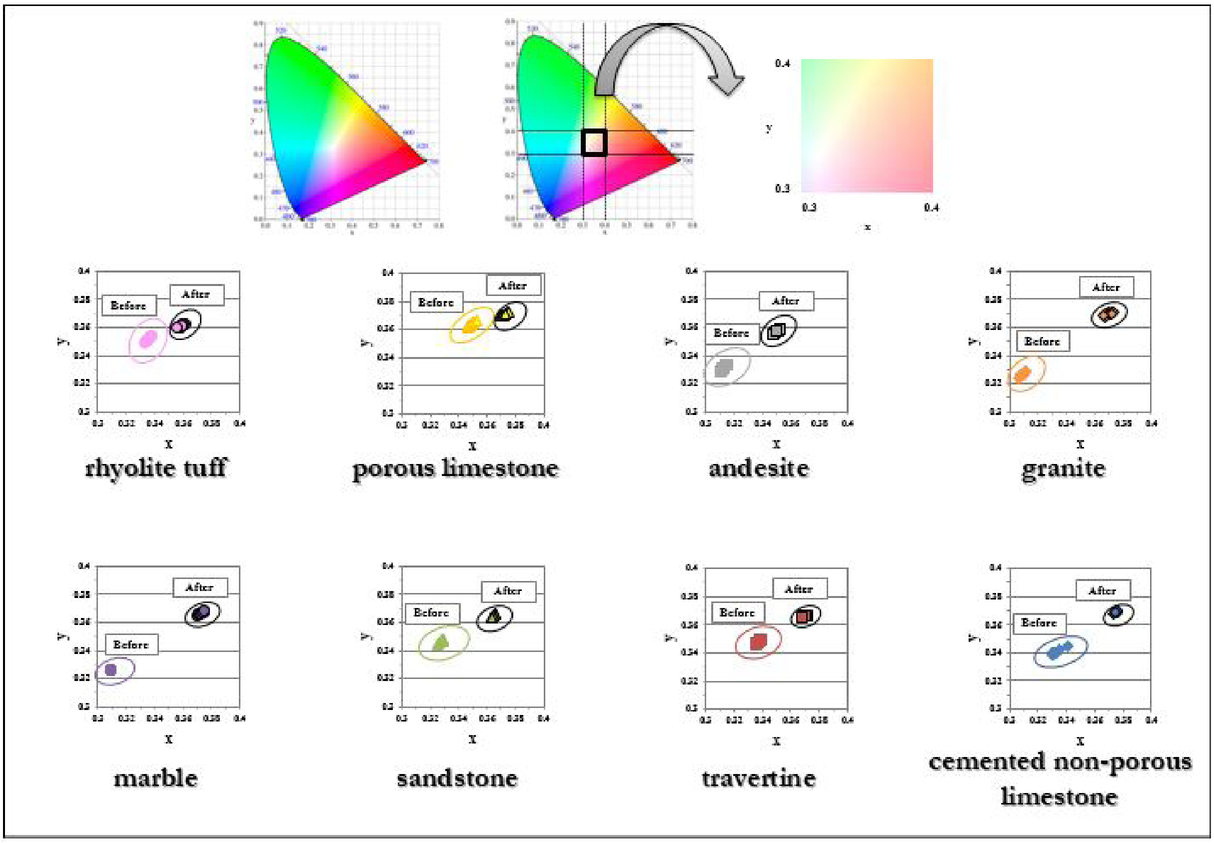

Fig. 5 The CIE 1931 colour space chromaticity diagram and the changes in the xy chromaticity coordinates of each tested stones (in coloured circles in the left side of the diagrams – chromacity before the test; in black circles in the right side of the diagrams – chromacity after the tets)

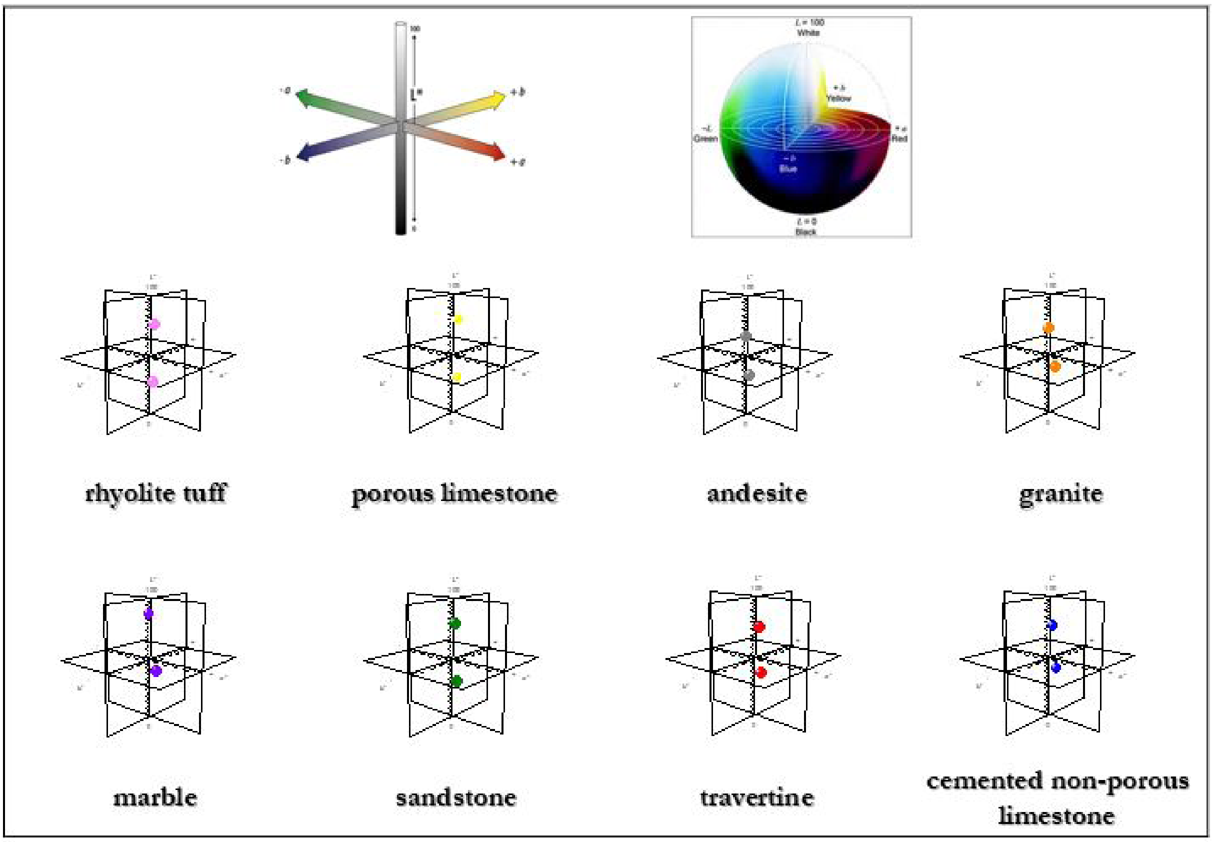

Fig. 6 The CIELAB colour space and the changes in the three (L*, a*, b*) coordinates of each tested stones (upper dots – before exposure, lower dots – after exposure)

space) coordinates (CIE document 15, 2004). The L*a*b* colour space describes all perceivable colours (Fig. 6).

After10 hours exposure the stone specimen's weight were reduced, particularly at the rhyolite tuff and andesite as shown by Fig. 7.

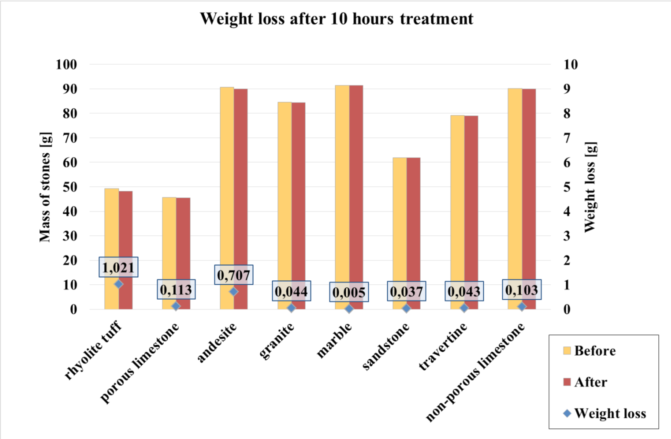

Fig. 7 Weight loss after 10 hours exposure

These weight loss may be related to high, ca. 250 °C temperature in the chamber and the differences of mineralogical composition (Török, 2003; Juhász and Kopecskó, 2013; Török and Török, 2015).

4 Conclusion

In order to investigate and determine under laboratory conditions the effect of ICE's exhaust gas on 8 different kinds of stones under laboratory conditions firstly a pollution process and after that a mass and colour measurements have been carried out. Our basic aim was to clarify and compare changes in the stones' colour.

The same trend was observed for all types of stone. Each coordinates are shifted in the direction of the yellowness, such as by Antal et al., 2017. Despite the short period of time of the test (10 hours) significant amount of particulate matter was deposited on stone specimens. A very high total colour difference value was recorded (Fig. 6). Our test has demonstrated colour changes, but this change may depend on the temperature, surface roughness and also on porosity, in addition to the amount of soot derived from the exhaust gas (Farkas et al., 2015; 2016)

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