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ScienceDirect

Transportation Research Procedia 45 (2020) 530–538

Transportation Research Procedia

www.elsevier.com/locate/procedia

AIIT 2nd International Congress on Transport Infrastructure and Systems in a changing world (TIS ROMA 2019), 23rd-24th September 2019, Rome, Italy

Durable Waterborne Horizontal Road Markings for Improvement of Air Quality

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Abstract

Horizontal road markings are very important safety features on almost all roads. For proper function, they must be reflectorised with glass beads, which provide visibility in the night time and simultaneously protect the paint from abrasion. Nonetheless, to maintain adequate performance frequent renewals are required. Field tests have demonstrated that durability achieved with modern waterborne road marking paints can be higher than measured with solventborne materials and that further improvements in durability of all paints can be achieved with the use of premium glass beads. Given that all paints dry because of evaporation of solvents to the atmosphere, emissions of Volatile Organic Compounds (VOC) reach approximately 25% for solventborne paints and below 5% for waterborne paints. The emitted VOC contribute to formation of ground-level ozone unequally, so Ozone Formation Potential (OFP) is used for a uniform comparison of various paints. For example, in Italy an annual reduction in OFP from 3,498 to only 43 tonnes could be realised by using highest quality waterborne road marking paints reflectorised with premium glass beads, due to the combined effects of lower individual OFP and improved durability. These results should be considered by road administrators and environmental agencies seeking a method to simultaneously improve air quality and decrease the frequency of renewals.

Peer-review under responsibility of the scientific committee of the Transport Infrastructure and Systems (TIS ROMA 2019).

Keywords: *horizontal road markings; road marking paints; retroreflectivity; tropospheric ozone; air pollution.*

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Peer-review under responsibility of the scientific committee of the Transport Infrastructure and Systems (TIS ROMA 2019).
10.1016/j.trpro.2020.03.060

1. Introduction

Since their introduction over a century ago, horizontal road markings became ubiquitous, essential safety features present on almost all modern roads. It was calculated that by organizing and channelling vehicular traffic the pavement markings bring sixtyfold financial savings over the total cost associated with their installation and maintenance (Miller, 1992). Vast majority of pavement markings are applied with waterborne or solventborne paints, because they are the least expensive initially, easiest to apply, and readily accessible worldwide (Babić et al., 2015). However, the application of paints is associated with the emissions of Volatile Organic Compounds (VOC), which are known to be harmful for the environment. VOC during decomposition process in the atmosphere can cause formation of tropospheric ozone in quantities depending on the selection of solvents. Based on a cradle-to-grave Life Cycle Assessment (LCA), it was calculated that the environmental sustainability of road markings depends not as much on the selection of paint or solvent, as on durability (i.e. service life) of the applied materials (Burghardt et al., 2016a, Cruz et al., 2016). Thus, increasing systems durability would decrease environmental burden.

This article is a continuation of the series of analyses addressing the potential of ground-level ozone formation from solvents used in horizontal road marking materials and the influence of durability on the VOC emissions. Herein, analysed are series of road marking events over the expected service life of road pavement, which is more applicable from the perspective of LCA of a road surface and accounts for the service life of the markings. Based on the results from field evaluation of road marking paints at urban and rural roads, it is shown that increase in the durability of pavement markings could be achieved by appropriate materials selection, which in consequence would meaningfully decrease the solvents contribution to formation of ground-level ozone, a key constituent of non-particulate smog. The provided analyses should be of particular importance to the environmental regulators and the road authorities in areas suffering from unacceptably high level of ozone pollution, such as the Mediterranean Sea basin.

2. Background

2.1. Horizontal road markings

Horizontal road marking materials are defined as systems comprising the colour layer (paint) and the retroreflective layer (glass beads) and only co-operation of these two layers furnishes a functional product (Pocock and Rhodes, 1952). Retroreflectivity of road marking, provided by glass beads, is absolutely necessary for driving during night time on unlit roads, when the number and quality of other visual cues are severely limited (Zwahlen and Schnell, 1999). Retroreflectivity is so critical for horizontal road markings that coefficient of retroreflected luminance (R_L) is used to evaluate the quality and functional life of pavement markings. While one could question the need for R_L in city environment where street lights provide sufficient illumination, it cannot be forgotten that glass beads layer also serves to protect the paint layer from abrasion and thus affect durability of the applied road marking system. Solvent-less road marking materials, which have different advantages and disadvantages, shall not be discussed in this article.

Road marking paints are composed of a binder (usually acrylic, but in very old formulations alkyd or chlorinated rubber binders can still be found), pigments and fillers, and additives – all suspended in a solvent (either an organic solvent or water or their mixture). Evaporation of the solvent after application makes the paint dry (in cause of modern waterborne paints, both an evaporation and a chemical reaction occur). Solvents constitute approximately 25% of a typical road marking paint (in case of waterborne paint, about 20% is water and VOC are limited to less than 5%). Considerable quantities of VOC are emitted every year to the atmosphere from road marking paints and their reduction would be advantageous to the society and the environment by minimising exposure to chemicals.

Durability of road marking paints was reported to vary from 6 to 24 months, depending on traffic load and type, marking location, weather conditions, and a plethora of other factors (Craig et al., 2007). More recently, field studies have shown that durability depends primarily on paint selection, but the choice of glass beads plays second very important role (Burghardt et al., 2016c). Premium glass beads, which are prepared in a proprietary process from virgin raw materials, can furnish not only high initial R_L, but also, due to their increased resistance to scratching, the highly desired longer service life of horizontal road marking systems (Burghardt et al., 2017; Burghardt, 2018; Burghardt et al., 2018). The selection of glass beads influences paint's functional life to the extent that it may in some cases bring long-term financial savings despite higher initial expense of purchasing premium materials (Burghardt et al., 2019). The premium glass beads have refractive index (RI) increased from the typical 1.5 to 1.6-1.7, but still belong to Class A according to the norm EN 1423:2012 (European Committee for Standardization, 2012), so there are no formal limitations for their utilisation in road markings required in vast majority of tenders. Such glass beads can be freely intermixed with standard glass beads.

2.2. Tropospheric ozone

While ozone layer in the stratosphere is beneficial because it protects the Earth from cosmic radiation, tropospheric ozone is simultaneously harmful and necessary. Ozone is a naturally-occurring allotrope of oxygen, responsible for self-cleaning of the atmosphere and playing very important role in the health and homeostasis of all ecosystems (Cape, 2008). However, since ozone is a very reactive species, it can cause severe irritation to respiratory system; in consequence, it was estimated that annually between 30 and 350 thousands of premature human deaths worldwide can attributed to the increased concentration of ground level ozone (OECD, 2012). Among statistical studies, in European countries even 140 Disability Adjusted Life Years per million inhabitants was reported to be lost because of ozone pollution (Hänninen et al., 2014) and deaths increase up to 2.9% was correlated with ozone concentrations rising by 50 $\mu g/m^3$ (Touloumi et al., 1997). Due to its high reactivity, ozone was also reported as causing damage to plants, thus influencing crops yields and quality (Fumagalli et al., 2001).

In clean, unpolluted air, ozone concentration is in an equilibrium with the level of nitrogen oxides (Chameides and Walker, 1973). However, all VOC are decomposing in the atmosphere *via* hydroxy radicals and they affect the equilibrium, which can lead to formation of additional ozone. Due to dissimilar chemical decomposition pathways, there can be a vast difference in reactivity of VOC and their potential of influencing ozone formation: some of the decomposing chemicals are re-introduced several times into the reactions, thus resulting in high quantities of produced ozone, while with a few others even ozone scavenging could occur (Crutzen, 1974). To quantify potential of tropospheric ozone formation from various solvents, a straightforward scale based on Maximum Incremental Reactivities (MIR) was developed (Carter, 1991). Instead of MIR, a Master Chemical Mechanism could also be utilised to estimate OFP with similar accuracy (Derwent et al., 2001). However, due to data availability for numerous chemicals, simplicity, and good application for high insolation areas like Mediterranean Sea basin the MIR scale was selected for calculations presented herein.

In climate like Mediterranean, summer weather conditions favour the formation of tropospheric ozone and disfavour its dissipation, which causes high ozone level episodes. They occur when stable high air pressure and anti-cyclonic circulation combined with weak winds cause the formation of offshore ozone reservoirs, which then move to the shore at daytime. Such ozone reservoirs can travel long distances (Monteiro et al., 2012). Quite interestingly, decade-long analysis of ozone concentration in Italy demonstrated that at rural measurement stations ozone concentrations were decreasing annually by an average of 0.43%, but simultaneously suburban and urban levels were annually rising, respectively by 0.46% and 0.64%. The increases in urbanised areas were attributed, paradoxically, to stricter vehicular emissions controls that lowered the availability of nitrogen oxides (Sicard et al., 2014). Therefore, limiting precursors of ozone formation, particularly in areas of high insolation, appears of critical importance.

2.3. Road marking paints and OFP

While there are quite strict regulations regarding VOC emissions from various types of consumer and industrial paints, those applying to road marking paints are scarce, if at all present. One of the key difficulties in estimating emissions are differences between countries in the definitions of VOC and the existence of 'exempt solvents' excluded from VOC content calculations. Therefore, for a unified comparison, OFP based on Total Organic Gases (TOG) emissions and MIR of the individual solvents can be conveniently utilised as was recently demonstrated (Burghardt and Pashkevich, 2018). Since TOG comprise all of the volatiles independent on their status and boiling point, they are more suitable for such calculations than the nominal VOC contents of a paint, which might be lower than actual emissions due to the regulatory assignments.

After a brief note showing the possibility of using OFP as a parameter in selecting road marking paints (Scorgie and Greenwood, 2011), an analysis was done on the examples of Kraków, Poland (Burghardt et al., 2016a) and the national roads network in Poland (Burghardt et al., 2016). A subsequent analysis demonstrated that in case of Croatia, a toluene-based road marking paint had OFP 27 times higher than waterborne road marking paint (Burghardt et al., 2016b). Summary of key findings from previous work is provided in Table 1. Whereas in previous work a one-time event of marking was analysed, in this article addressed shall be OFP of exemplary paints calculated as markings renewals required during the usual life span of the roadway surface.

Table 1. Previous analyses.

Article	Analysed region	Key findings
Scorgie and Greenwood, 2011	New Zealand	Not only VOC contents, but also OFP can be used for assessment of environmental friendliness of road marking paints.
Burghardt et al., 2016a	Kraków, Poland	Up to 43 tonnes of tropospheric ozone can be produced annually from solvents emitted from road markings used in Kraków. Annual reduction of approximately 39 tonnes of ozone could be realised by a switch to waterborne paints.
Burghardt et al., 2016	Poland	About 500 tonnes of road marking paints used annually to mark national roads network in Poland can produce over 1,000 tonnes of tropospheric ozone, which is due to the use of a paint containing up to 8% of toluene.
Burghardt et al., 2016b	Croatia, Italy	There is about a tenfold difference in OFP between typical various organic solvents used in road marking paints. Solvents evaporating from 1.0 kg of road marking paint can produce between 0.036 kg (waterborne paint) and 0.958 kg (toluene-based solventborne paint) of tropospheric ozone.
Burghardt and Pashkevich, 2018	United States, Croatia, Poland, Austria, Sweden	Significant incongruity between the declared VOC emissions and OFP was calculated in case of some low VOC paints based on ‘exempt solvents’. OFP based on TOG is better suitable for estimation of environmental friendliness and limiting emissions than VOC-based regulations because it accounts for all volatile compounds regardless of their legislative status.

3. Methodology

Whereas theoretical calculations of TOG emissions and OFP can be easily done by analysing composition of road marking paints’ volatile portions, the results are applicable to only one event of road marking. However, such activities are done periodically and as such it appears more appropriate to treat them as a series of events associated with the service life of the road surface, which in this case is assumed as 20 years. No articles addressing the consumption of road marking materials and the associated emissions over a service life of a road surface were found, except the aforementioned LCA analyses (Cruz et al., 2016; Burghardt et al., 2016a). Herein, for OFP calculations previously established methodology is being utilised (Burghardt and Pashkevich, 2018). Then, based on durabilities of the evaluated road marking systems, TOG emissions and the resulting OFP per a 20-year cycle for selected road marking systems is assessed. Durability (service life of a road marking) is obtained from field tests of various road marking systems done under dissimilar conditions, which demonstrate that the selected materials behave similarly, even if their service life is shorter under heavy city traffic. For service life estimation, an exponential line fit is used because it gave the most realistic results in similar work (Burghardt et al., 2017; Burghardt et al., 2018; Burghardt, 2018).

The field testing of paint-based road marking systems were done in Croatia (rural environment, at a major two-lane road; line markings 15 cm wide) and in Poland (urban environment, in the city of Kraków, at an intersection approach of a feeder road with 2+1 lane arrangement; line markings 12 cm wide). The applications were done by local crews, using their standard protocols and equipment. All of the tested paints were applied at wet film thickness of 350-450 µm (500-600 g/m²) and were immediately reflectorised with drop-on glass beads, injected under mild pressure at 350-400 g/m². In both cases, the solventborne paints (toluene-based in Croatia and toluene-containing in Poland) and standard glass beads with RI 1.5 and roundness >80% were provided by the application crews; these were materials that they used during regular work. Modern waterborne quick-dry road marking paint Limboroute® W13 (Swarco Limburger Lackfabrik GmbH; Diez, Germany) and premium glass beads (RI increased to 1.6-1.7, roundness >90%, and improved resistance to scratching) SOLIDPLUS (M. Swarovski GmbH; Amstetten, Austria) were furnished by their manufacturers as free of charge professional samples.

Road markings quality and service life is customarily measured by R_L and this method was employed here. Measurements of R_L were done with a portable retroreflectometer calibrated for the typically used 30-metre geometry; 7 data points were collected per line (always in the same location). Due to various local limitations a full testing matrix could not be applied; hence, only limited information can be provided. For the calculations of long-term TOG emissions and OFP, it is assumed that renewal of road markings is done when R_L drops to 100 mcd/m²/lx, which is the typical minimum required by road administrators, particularly for city environment. It ought to be noted that R_L>150 mcd/m²/lx is currently recommended by European Road Federation and was suggested after assessing visual needs assessment of general population (Gibbons et al., 2012).

4. Results

4.1. Durability of road marking systems

The collected results from the field tests are provided in Table 2. Improved performance achieved with premium glass beads, even if they were only intermixed with standard beads, is obvious. In other work (Burghardt et al., 2016c; Burghardt et al., 2019) it was demonstrated that the use of premium glass beads with solventborne road marking paint causes excellent initial R_L, but durability is not improved meaningfully; therefore, solventborne paint was not reflectorised with premium glass beads for the purpose of these experiments. The increase in service life was measured only with the use of waterborne paints combined with the use of premium glass beads. One should note that different line location, different traffic load, and different weather conditions make a just side-by-side comparison impossible and consider the provided information only as an indication. Urban environment in Poland and higher traffic load definitely negatively affected the service life of the applied markings.

Table 2. Applications in Croatia (rural) and in Poland (urban).

Country	Croatia (rural environment)				Poland (urban environment)			
Paint[(a)]		SB (T)	WB	WB		SB (TC)	WB	WB
Glass beads		Standard	30% premium	100% premium		Standard	30% premium	100% premium
Line location		Edge	Centre	Centre		Centre	Middle[(b)]	Centre
	Traffic[(c)]	R_L [mcd/m²/lx][(d)]			Traffic[(c)]	R_L [mcd/m²/lx][(d)]		
Initial	0.1	483 (26)	645 (22)	677 (12)	0.2	309 (9)	325 (43)	631 (28)
After winter	1.4	303 (31)	558 (4)	541 (56)	2.1	138 (5)	254 (38)	403 (42)
After two winters	2.7	253 (12)	330 (4)	437 (70)	–	n/a	n/a	n/a
Durability estimate [years]; R_L<100 mcd/m²/lx[(e)]		4	5	7		1	3	3.5

[(a)]SB(T) – solventborne toluene-based paint, SB(TC) – solventborne toluene-containing paint, WB – waterborne paint. [(b)]Marking dividing lanes in the same direction. [(c)]Traffic load provided in millions of vehicle passes in one direction, per data obtained from the local road authorities and assuming equal traffic in both direction. [(d)]Average R_L, standard deviations provided in parentheses. [(e)]Estimate based on exponential line fit.

4.2. VOC emissions and OFP

Volatile portions of selected commonly used road marking paints were collected based on publicly-accessible Safety Data Sheets. In case of waterborne paint, the composition was supplemented with a typical coalescent (mixed isomers of 2,2,4-trimethyl-1,3-pentanediol monoisobutyrate, TMPDMIB) and water. Knowing MIR of the solvents

(California Code of Regulations, 2015), it was possible to calculate the OFP of these paints. Data is furnished in Table 3. It ought to be noted that particularly high MIR of toluene is making containing it paint less environmentally friendly than an aromatic-free paint in terms of OFP. Waterborne paint is obviously contributing least to the formation of tropospheric ozone.

Table 3. Composition of volatile portions of exemplary road marking paints and the resulting OFP.

Solvent (VOC)	MIR	Toluene-based	Toluene-containing	Aromatic-free	Waterborne
Toluene	4.00	24.3%	8.0%	–	–
Butyl acetate	0.83	–	–	20.0%	–
Ethyl acetate	0.63	–	6.0%	–	–
Methyl ethyl ketoxime	1.58	0.2%	–	–	–
4-methyl-2-pentanone	3.88	–	10.0%	–	–
Butan-2-one	1.48	–	–	5.0%	–
TMPDMIB	0.81	–	–	–	1.8%
Ethanol	1.53	–	–	–	0.4%
Ammonia	–[(a)]	–	–	–	0.3%
Water	–[(a)]	–	–	–	21.6%
VOC[(b)]		24.5%	24.0%	25.0%	0.7%
TOG		24.5%	24.0%	25.0%	2.5%
Ozone Formation Potential (OFP) [g O_3 / kg paint]		975	746	240	21

[(a)]Value not established or zero. [(b)]Based on VOC definitions and regulations in European Union.

Whereas data provided in Table 3 gives information for an individual paint and a one-time event of road marking, in Table 4 are provided long-term emissions for various renewal frequency, with the durability of road marking taken into account. The durability was obtained from Table 2, based on the results from the test in urban environment in Poland. In all of the cases, calculations were done for paint applied at 600 g/m² wet film. It is assumed that durability of solventborne road marking paints would be equal, independent on their solvent composition, which might be incorrect in some cases (Burghardt et al., 2016).

Table 4. Long-term emissions from various road marking systems.

Paint type	Solventborne, toluene-based	Solventborne, toluene-containing	Solventborne, aromatic-free	Waterborne	
Paint VOC[(a)]	25%	24%	25%	0.7%	
Paint TOG	25%	24%	25%	2.5%	
OFP [kg O_3/kg paint]	0.975	0.746	0.240	0.021	
Glass beads	Standard	Standard	Standard	30% premium	100% premium
Durability estimate [years]	1	1	1	3	3.5
Renewals per 20 years	20	20	20	7	5
TOG [kg/m²]	0.15	0.14	0.15	0.015	0.015
TOG per 20 years [kg/m²]	2.94	2.88	3.00	0.105	0.075
OFP [kg O_3/m²]	0.59	0.45	0.14	0.012	0.012
OFP per 20 years [kg O_3/m²]	11.70	8.95	2.88	0.087	0.062

[(a)]Based on VOC definitions and regulations in European Union.

4.3. Reduction in OFP scenario

Based on data provided in Table 4, while knowing the most frequently used type of road marking paints in different countries, one could estimate the annual OFP and possible environmental benefit if a switch to a modern waterborne paint occurred. Simulation data for these scenarios is given in Table 5 for selected European countries. Estimates for domestic consumption in 2006 were obtained from an industry report (Ökopol, 2011). An OFP reduction scenarios with a switch to waterborne road markings is provided in Table 5, with and without the assumption of increased service life (achieved with the waterborne paint reflectorised with premium glass beads in the urban environment).

Table 5. Estimated annual usage of road marking paints in 2006 and associated emissions.

Country	Annual usage [tonnes]	Solventborne share	Type of solventborne paint	Annual TOG emissions [tonnes][a]	Annual OFP [tonnes O_3][a]	Switch to waterborne OFP [tonnes O_3][b]	Minimum OFP [tonnes O_3][c]
Austria	2,001	79%	Aromatic-free	405	389	41 (-89%)	12
Croatia	1,069	100%	Toluene-based	262	1,042	22 (-98%)	6
Italy	7,191	79%	Toluene-based 50%	1,440	3,489	149 (-96%)	43
			Aromatic-free 50%				
Poland	16,393	100%	Toluene-containing	3,934	12,226	339 (-97%)	97
Spain	12,061	79%	Aromatic-free	2,438	2,343	250 (-89%)	71
Sweden	970	7%	Aromatic-free	36	35	20 (-43%)	6

[a] Includes the contribution from waterborne paints. [b] Assuming annual renewal. [c] Assuming renewal every 3.5 years, which could be achieved with 100% premium glass beads (Cf. Table 2).

5. Discussion and Conclusions

The estimates provided herein, based on field tests, demonstrate that a switch to a modern waterborne road marking paints reflectorised with premium glass beads would more than triple the life span of the pavement markings. Consequently, the reductions in VOC emissions and OFP, already minimised by the use of a waterborne paint, would be further meaningfully depressed: from a maximum calculated 11.70 kg of O_3 (solventborne toluene-based paint, with durability of 1 year in urban environment) to 0.06 kg of O_3 (waterborne paint reflectorised with premium glass beads, with durability of 3.5 years in urban environment) per 1 m² in the 20-year period. If the same scenario were applied to individual countries, in Italy a possible reduction in the potentially formed tropospheric ozone from 3,498 tonnes to 43 tonnes annually could be realised with simultaneous materials consumption lowered by an estimated 2,054 tonnes annually. A reduction in OFP could exceed 99% for a country using solely solventborne toluene-based paint. Nonetheless, one must observe that a complete switch to waterborne road markings in all countries is unlikely because of specific local circumstances, particularly those associated with weather. Nonetheless, a very meaningful reduction in both OFP and consumption of materials can be achieved and ought to be pursued because of possible significant environmental advantage.

Reduction in tropospheric ozone can be converted into improved health of the population (Touloumi et al., 1997; OECD, 2012; Hänninen et al., 2014). Even if solvents emitted from road marking paints are quite a small contributor to the overall tropospheric ozone production, their choices that are less harmful are readily available. Although the premium-quality materials are more expensive, their costs can be offset by the environmental benefits and longer service life. In addition, it must be kept in mind that financial spending on improvement of infrastructure was correlated with lower accident rates (Albalate et al., 2013), which would be an additional benefit. It was recently shown that in the cities there is a positive correlation between the quality of road markings and road safety (Diependaele, 2018). Furthermore, lesser renewal frequency required with the described premium products could additionally lead to lower traffic congestion (Fiolić et al., 2017).

In conclusion, it was demonstrated that various road marking paints with the same nominal VOC contents may vary very significantly if one uses OFP of TOG to compare them. The lowest VOC emissions and lowest OFP were calculated for a waterborne paint; the only impactors there were the coalescent and ethanol. In addition, because modern waterborne paints, especially in combination with premium glass beads, were measured to be more durable than solventborne systems, very significant further lowering of emissions can be realised. Therefore, the most environmentally-friendly choice would be the use of a waterborne paint reflectorised with appropriately matched premium glass beads. Such option should be considered not only by the road administrators, but also requested by environmental protection authorities.

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