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REDUCTION OF PARTICULATE MATTER EMISSIONS IN EU INLAND WATERWAY TRANSPORT

Master's thesis for the degree of Master of Science in Technology submitted for inspection, Vaasa, 22 April, 2016.

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CONTENTS

LIST OF ABBREVIATIONS

CARB	California Air Resources Board
CCRT	catalysed continuously regenerating technology
CDPF	catalysed diesel particulate filter
CO	carbon monoxide
CRT	continuously regenerating technology
DOC	diesel oxidation catalyst
DPF	diesel particulate filter
DPM	diesel particulate matter
EPA	United States Environmental Protection Agency
FBC	fuel borne catalyst
HC	hydrocarbons
IWT	inland waterway transport
NRMM	non-road mobile machinery
NRSC	Non-Road Steady Cycle
NRTC	Non-Road Transient Cycle
NTP	non-thermal plasma
PAH	polycyclic aromatic hydrocarbons
PGM	platinum group metal
PM	particulate matter or particulate mass
$PM_{0.1}$	ultra-fine particles 50–100 nm
$PM_{2.5}$	fine particles 0.1–2.5 µm
PM_{10}	coarse particles 2.5–10 µm
PN	particulate number
POC	particle oxidation catalyst
ppm-wt.	parts per million by weight
SCR	selective catalytic reduction
SDPF	diesel particulate filter with SCR coating

SiC	silicon carbide
SO_4	sulphate (particulates)
SOF	soluble organic fraction of diesel particulates
SOL	solid fraction of diesel particulates
TPM	total particulate matter
ULSD	ultra-low sulphur diesel
UTAC	United Test and Assembly Center
VERT	Verification of Emission Reduction Technologies
VOC	volatile organic compound
WES	wet electrostatic scrubber
WHTC	World Harmonized Transient Cycle

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ABSTRACT

In September 2014, the European Commission adopted a proposal on new requirements relating to emission limits and type-approval for non-road engines. The introduction of a new emission stage (Stage V) establishes extremely tight limits for particulate matter emissions for mobile non-road applications, including inland waterway vessels. These new emission limits will eventually require many ships to apply efficient exhaust gas after-treatment technology.

The aim of this study was to find out which kinds of exhaust gas after-treatment solutions could fulfil these tightening particulate emission standards in EU inland navigation. A marine dual fuel engine was used as an example. The engine can be run both with gas and diesel fuel.

The first part of the study consists of a literature review of various exhaust gas after-treatment technologies. This part serves as a general technology guide for particulate emission abatement from diesel engines. In the second part of the study, different supplier technologies and solutions were evaluated. The targets for particulate filtering system were defined and a specific inquiry was sent to potential suppliers. Based on the replies, passive diesel particulate filter systems with catalytic coating or/and an upstream diesel oxidation catalyst can be regarded as the primary choice for particulate emission control in inland navigation.

This study was conducted as part of the EU Hercules-2 research and development programme, aimed at fostering environmentally sustainable and more efficient shipping.

KEYWORDS: inland waterway transport, gas and diesel engines, particulate matter emissions, diesel particulate filter

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TIIVISTELMÄ

Euroopan komissio antoi syyskuussa 2014 ehdotuksen uusista työkonemoottoreiden päästöraja-arvoihin ja tyyppihyväksyntään liittyvistä vaatimuksista. Ehdotuksella tiukennetaan liikkuvien työkoneiden, mukaan lukien sisävesialusten polttomoottoreiden pakokaasupäästöraja-arvoja nykyisestä ottamalla käyttöön uusi vaihe V. Ehdotettujen erittäin tiukkojen hiukkaslukumäärävaatimusten noudattaminen edellyttää jatkossa alusten moottoreiden varustamista tehokkaalla pakokaasujen jälkikäsittelytekniikalla.

Tutkimuksessa selvitettiin, minkälaisin pakokaasujen jälkikäsittelyratkaisuin laivamoottori voidaan saada täyttämään nämä EU:n sisävesille tulevat vaativat hiukkaspäästönormit. Esimerkkinä käytettiin monipolttoainemoottoria, jota voidaan ajaa niin kaasu- kuin dieselpolttoaineella.

Soveltuvia pakokaasujen jälkikäsittelyratkaisuja koskeva kirjallisuuskatsaus muodostaa työn ensimmäisen osan. Lisäksi tämä osa kuvaa yleisluontoisesti dieselmoottoreiden hiukkaspäästöjen hallintaan soveltuvia erilaisia jälkikäsittelytekniikoita. Työn toisessa osassa arvoitiin eri laitetoimittajien markkinoimia tuotteita ja ratkaisuja ja niiden soveltuvuutta monipolttoainemoottoreihin. Jälkikäsittelyjärjestelmää koskevat tavoitteet määriteltiin, ja yksityiskohtainen kysely lähetettiin usealle alan toimijalle. Vastausten perusteella passiivisesti regeneroituvia dieselhiukkassuodinjärjestelmiä voidaan pitää ensisijaisena valintana sisävesialusten hiukkaspäästöjen hallintaan.

Tutkimus kuuluu Euroopan unionin rahoittamaan Hercules-2 -tutkimus- ja kehitysohjelmaan. Ohjelmassa kehitetään ympäristön kannalta kestävää ja tehokasta merenkulkua.

AVAINSANAT: sisävesiliikenne, diesel-ja kaasumoottorit, pienhiukkaspäästöt, hiukkassuodin

1 INTRODUCTION

Inland waterway transport (IWT) has been the most environmentally friendly mode of inland transport for decades. However, this advantage has steadily been undermined due to the rapid improvement of emissions in the road transport sector. In contrast to the road transport sector, the replacement rate of engines used in inland waterway vessels is very low and the emission standards for new engines have been much less strict. As a consequence, inland waterway transport for certain routes, cargo types and vessel sizes already has higher air pollutant emission levels than road transport per tonne kilometre. Without specific action on the legislative field, the traditional environmental advantages of IWT will further deteriorate in the future. (Panteia 2013: 17.)

Cargo vessels on inland waterways are typically equipped with internal combustion engines burning diesel oil. In contrast to maritime navigation, the legally acceptable sulphur content of inland navigation fuels in the EU is so low that these fuels can be seen as quasi sulphur free. Therefore, nitrogen oxides (NO_x), particulate matter (PM), hydrocarbons (HC) and carbon monoxide (CO) constitute the main pollutant emissions from engines for inland navigation. (Pauli 2016: 485.) Among these, in particular diesel PM has been associated with adverse health effects in humans and is classified as a human carcinogen. One of the main findings is that the size of the particles is a crucial factor behind the observed health effects (EU Proposal 2014/0581). In addition, the strongly light absorbing fraction of PM, black carbon, has been identified as an important climate forcer with a high global warming potential (Yelverton et al. 2015).

Diesel particulates are subject to diesel emission regulations worldwide. The first European Union wide compulsory emission limits for inland waterway vessels were introduced with Directive 97/68/EC on non-road mobile machinery, which has applied to new vessels since 2004. The currently applicable Stage IIIA standards within this framework are now under revision. (Panteia 2013: 19.) In September 2014, the European Commission proposed a new regulation on requirements relating to emission limits and type-approval for internal combustion engines for non-road mobile

machinery, including inland navigation vessels. The proposal includes the introduction of a new emission stage (Stage V). This stage includes a number of important changes. It widens the scope of regulated engines, strengthens the emission limits, and furthermore, adopts particle number (PN) emission limits for several categories. (DieselNet 2105a.) The particulate number can be seen as a tool to limit ultrafine particulates from diesel engines, which are known to play the largest role in affecting human health (Pauli 2016: 511).

The new type approval dates are expected to enter into force gradually from 2018 to 2020. After this, new emission limits will eventually require many ships to apply an efficient exhaust gas after-treatment technology. The objective of this study was to explore available after-treatment methods for the removal of exhaust PM. The main target was to research with which kinds of exhaust gas after-treatment solutions a marine dual fuel engine could fulfil the future tightening PM emission standards in inland waterway transport in Europe, both in gas and diesel fuel use.

A literature review was conducted. A number of PM reduction system suppliers were contacted, and an inquiry was sent to several manufacturers concerning a PM removal system for 1 MW medium-speed marine dual fuel engine. Proposals for particulate filtering system were received from several suppliers. Based on the replies, passive diesel particulate filter systems with catalytic coating or/and an upstream diesel oxidation catalyst can be regarded as the primary choice for particulate matter emission control in IWT applications.

This study is organized as follows. The research methods are described in Chapter 2. Chapter 3 focuses on the theoretical background of particulate matter, the process of formation of diesel particulate matter in combustion engines and the composition and structure of diesel particulates. The particle size distribution is also discussed, and a review of the effects of diesel PM on human health is made. In Chapter 4, the emission standards for inland waterway transport in European Union are introduced. After-treatment technologies for particulate matter reduction are described in Chapters 5–7.

The pros and cons of different technologies are evaluated, and special attention is paid on emission reduction efficiencies, regeneration methods and maintenance requirements. Among these techniques, diesel particulate filters (DPFs) with high filtration efficiency and good regeneration characteristics were considered as the best option. In Chapter 8, an evaluation of different supplier technologies and solutions is made. Targets for particulate filtering systems were defined and a specific inquiry was sent to potential suppliers. The replies are outlined and compared, and finally, the main findings and general conclusions are summarized in Chapter 9.

This study was conducted as a part of the Hercules-2 research and development programme, aimed at fostering environmentally sustainable and more efficient shipping. The Hercules-2 project is in line with general European Union policy and is mainly funded by the EU.

2 RESEARCH METHODS

In the first part of this study, a literature review was conducted. This first part serves as a general description of different exhaust gas after-treatment technologies for particulate emission control, including catalytic converters, particulate filters, and auxiliary systems for supporting particulate filter regeneration. In addition to these conventional techniques, integrated systems for simultaneous control of several air pollutants were overviewed.

In the second part of the study, an evaluation of different supplier technologies and solution was made. The aim was to find an effective and reliable particulate filtering system for 1 MW marine dual fuel engine, both in gas and in diesel fuel use. The efficiency of the system had to be high enough to meet the PM emission limits specified in the proposed EU Stage V emission standards for inland waterway transport.

In order to find an optimal after-treatment system for the 1 MW dual fuel engine, several companies active within the exhaust gas after-treatment field were reviewed. The particulate filtering suppliers were found by surveying literature and the Internet. A specific inquiry was sent to potential suppliers. The inquiry contained information about the targets for particulate filtering systems, engine data and information about the fuels and the exhaust gas parameters. The suppliers were asked to specify the following items in their proposals: design and size of the system, PM and PN removal efficiency, pressure drop over the filtering system, soot regeneration method, maintenance requirements and expected ash cleaning interval, warranty terms, price and delivery time.

3 PARTICULATE MATTER THEORY

In an ideal combustion process, all the fuel injected to the engine is burned, leaving behind only carbon dioxide and water. However, combustion in internal combustion engines is never complete. (Pihlava, Uuppo & Niemi 2013: 8.) In incomplete combustion, products from partial combustion and pyrolysis and part of the unburned fuel contaminate the flue gas with noxious particulate matter. Diesel particulate matter is a complex mixture of organic and elemental carbon, acids, such as sulphates and nitrates, metals, particle-bound water and soluble organic compounds, such as PAHs and dioxins. (Majewski & Khair 2006: 105, 131-132; WHO 2013: 2.) In this chapter, the process of formation of diesel particulate matter in combustion engines as well as the composition and structure of diesel particulates and particle size distribution are discussed. Furthermore, a review of the effects on human health of diesel PM is made.

3.1 Particle Composition and Structure

In order to take steps to restrict the emission of particulate matter, it is necessary to identify and describe the mechanism leading to PM formation during engine operation. (Merkisz & Pielecha 2015: 20.)

PM is an extremely complex mixture of solid and condensates materials. The primary materials found in the solid phase of diesel PM include elemental carbon and metal ashes, while condensates include high boiling hydrocarbons, water and sulphur acid. (Guan & al. 2015.) The typical chemical composition and structure of diesel engine exhaust particulate matter is shown in Figure 1.

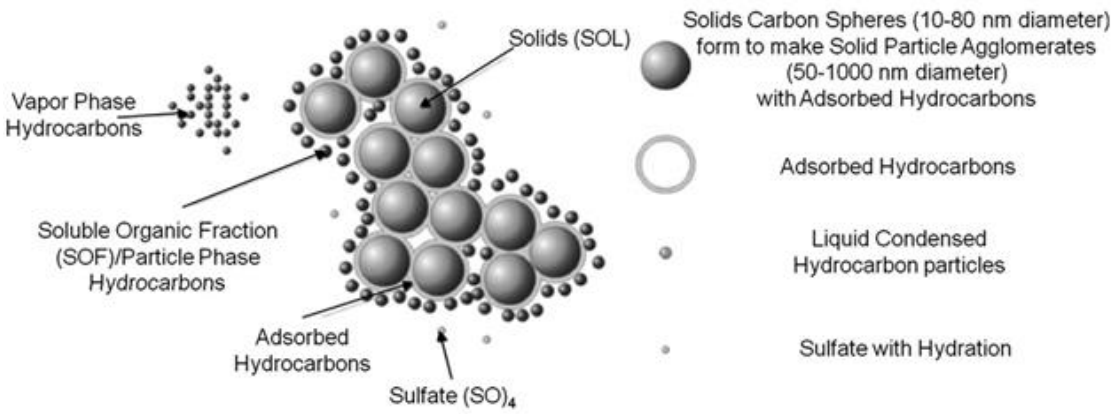

Figure 1. Typical chemical composition and structure of diesel PM (Guan 2015).

Particulate matter is traditionally divided into three main fractions, which can be further subcategorized as follows (Majewski & Khair 2006: 127-128):

1. Solid fraction (SOL)
   - elemental carbon
   - ash
2. Soluble organic fraction (SOF)
   - organic material derived from engine lubricating oil
   - organic material derived from fuel
3. Sulphate particulates (SO_4)
   - sulphur acid
   - water

Thus, the total particulate matter (TPM) can be defined as:

$$TPM = SOL + SOF + SO_4 \quad (1)$$

The composition of PM varies to a great extent depending on the engine technology, engine load, and, in the case of sulphate particulate, the sulphur content in the fuel. The typical PM composition from a heavy-duty diesel engine is illustrated in Figure 2.

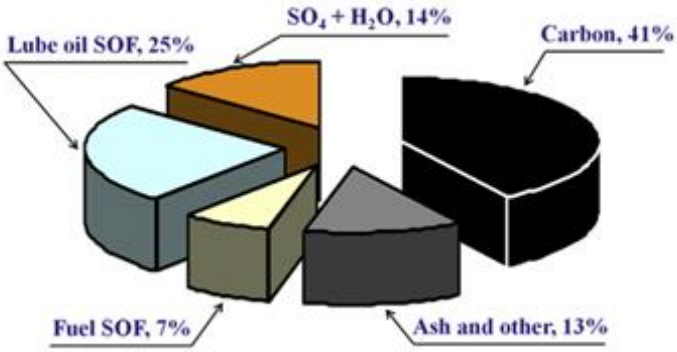

Figure 2. Composition of diesel particulate matter for an HD diesel engine (Guan & al. 2015).

3.1.1 Solid Fraction

The solid fraction of diesel particulates (SOL) is composed primarily of elemental carbon. This carbon, not chemically bound with other elements, is the finely dispersed "carbon black" or "soot" substance responsible for black smoke emissions. (Majewski & Khair 2006: 128.) Solid carbon is formed during combustion in locally rich regions. Much of it is subsequently oxidized. The residue is exhausted in the form of solid agglomerates, i.e. clusters, formed from primary particles. In addition, metal compounds in the fuel and lube oil lead to a small amount of inorganic ash. (Kittelson 1998.)

In diesel engines soot formation starts as a result of the oxidation of fuel molecules and/or thermal decomposition of unsaturated hydrocarbons, including acetylene and its derivatives, and polycyclic aromatic hydrocarbons (PAH). When particle condensation takes place, the first identifiable soot particles, called nuclei, occur. These tiny particles, with a diameter of about 2 nm, are very mobile and collide with each other; as a result, large structures with the number of atoms higher than 10^5 are formed. Their aerodynamic diameter (diameter of the spherical particle with a density of 1 g/cm^3 that

has the same settling velocity as the particle of the interest) ranges from 10 to 80 nm, but particles with dimensions of 15–30 nm are most common. The main factors affecting the formation of soot are fuel parameters, the fuel injection process, combustion pressure and combustion chamber shape. (Merkisz & Pielecha 2015: 20.)

After nucleation, an increase of the surface area of soot particles and agglomeration takes place. The surface growth follows from the combination of particles with those already existing through nucleation and condensation. Most of the mass is added during this phase and the residence time has a large impact on the total soot mass that is created. Agglomeration is the process of colliding particles, due to which a smaller number of larger particles are created. (Dembinski 2014: 10; Merkisz & Pielecha 2015: 20.) The different steps of soot formation are shown in Figure 3.

Both individual (nuclei mode) and agglomerated carbon particles are formed in the combustion chamber. The primary particles agglomerate in the cylinder while traveling through the exhaust system, and after discharge into the atmosphere. (Majewski & Khair 2006: 129.)

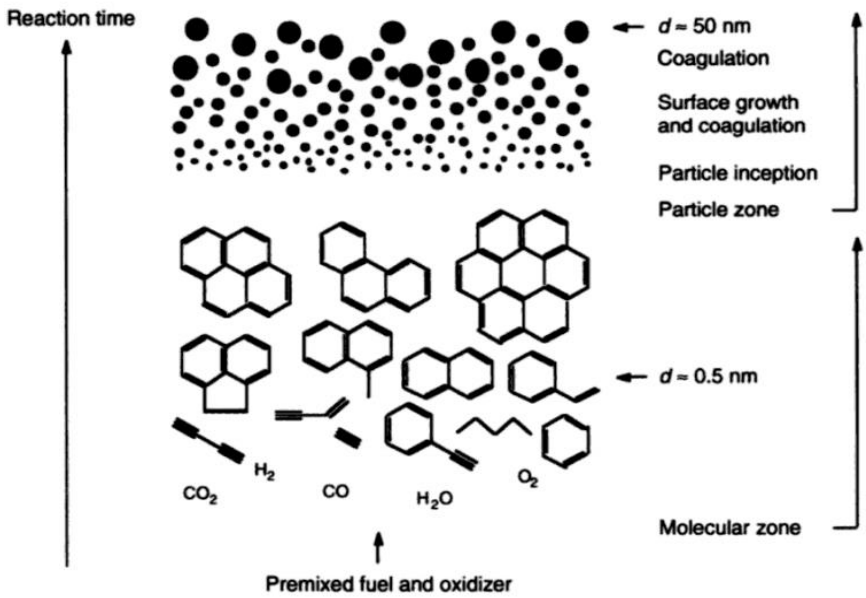

Figure 3. Schematic plot of soot formation in combustion (Dembinski 2014).

Another main component of the solid fraction of PM is metallic ash. In general, diesel exhaust ash consists of a mixture of the following components:

- Sulphates, phosphates, oxides of calcium, zinc, magnesium and other metals that are formed in the engine's combustion chamber from burning of additives in the engine lubricating oil.
- Metal oxide impurities resulting from engine wear, which are carried into the combustion chamber by the lube oil. These include oxides of iron, copper, chromium and aluminium.
- Iron-containing oxides resulting from corrosion of the engine exhaust manifold and other exhaust system components. (Merkel et al. 2001.)

Metal oxides can assume a significant proportion of the particulate mass when an additive is blended into fuel for particulate filter regeneration (Tschöke 2010: 447).

3.1.2 Soluble Organic Fraction

A tiny fraction of the fuel and atomized and evaporated lube oil escapes oxidation and appears as soluble organic compounds (SOF) in the exhaust (Kittelson 1998). At the temperature of diesel exhaust, most SOF compounds exist as vapours, especially at higher engine loads when the temperature is high (Majewski & Khair 2006: 130). When temperature decreases, vaporized hydrocarbons condense on the surface of soot particles partly in the exhaust pipe, but also when they have reached the air. Gaseous compounds comprise new, different chemical compounds when they react together and with compounds found in the air. At this point, when the exhaust gas reaches the outdoor air and the temperature decreases, most ultra-fine particles (diameter smaller than 0.1 µm) are formed and gaseous hydrocarbons start to condense into small particle droplets. (Pihlava et al. 2013: 8-9.)

Soluble organic fraction is typically composed of lube oil derived hydrocarbons, with a small contribution from the higher boiling end diesel fuel hydrocarbons. The most

harmful hydrocarbon compounds in diesel PM are polycyclic aromatic hydrocarbons (PAHs) and dioxins. PAHs are aromatic hydrocarbons with two or more benzene rings joined in various, more or less clustered forms. They may also contain cyclopentane rings and heterogeneous rings with atoms of nitrogen or sulphur. PAHs have attracted special attention because of their mutagenic and carcinogenic character. (Majewski & Khair 2006: 130-131.)

PAHs are present in diesel fuel. It is also believed that some of the heaviest PAH compounds are generated by pyro-synthesis in the engine cylinder. Emissions of PAHs typically constitute a fraction of a percent of total PM emissions, with many studies reporting about 0.5 % of total PM (Rogge et al. 1993).

Dioxin is the generic term for a special group of chlorinated polynuclear hydrocarbon compounds characterized by extremely high toxicity, suspected carcinogenicity, and resistance to biological breakdown. Certain catalytic combustion additives may increase emissions of dioxins by orders of magnitude. Therefore, fuel additives must always be evaluated for their dioxin formation activity. (Majewski & Khair 2006: 132-133.)

3.1.3 Sulphate Particulates

Sulphate particulates (SO_4) are composed primarily of hydrated sulphuric acid and, as such, are mostly liquid (Majewski & Khair 2006: 133). The particulates' sulphate fraction is basically derived from sulphur compounds in the fuel and to a lesser extent in the engine oil. During combustion, the sulphur oxidizes into SO_2 and, at exhaust gas temperatures above 450˚C, into SO_3. Interaction with water causes the formation of sulphate ions SO_4^{2-} to produce sulphuric acid H_2SO_4. (Tschöke et al 2010: 447.) When the exhaust gases are discharged from the tailpipe and mixed with air, their temperature decreases. Under such conditions the gaseous H_2SO_4 combines with water molecules and nucleates, forming liquid particles composed of hydrated sulphuric acid. (Majewski 2015a.)

In addition to sulphuric acid, sulphate particulates may also include sulphate salts. The most common salt is calcium sulphate $CaSO_4$, which can be formed in reactions between H_2SO_4 and calcium compounds originating from lube oil additives. Various sulphates may be also produced in reactions between sulphuric acid and exhaust system components. It is believed that sulphate particulates are separate from carbon particles and are present in the exhaust gas primarily as nuclei mode particles. (Majewski & Khair 2006: 133.)

3.2 Particle Size

Guan & al. (2015) divide ambient particulate matter into the following five categories based on their aerodynamic diameter:

1. large particles >10 µm
2. coarse particles 2,5-10 µm (PM_{10})
3. fine particles 0,1-2,5 µm ($PM_{2.5}$)
4. ultra-fine particles 50-100 nm (0,1 µm) ($PM_{0.1}$)
5. nanoparticles <50 nm

Nearly all diesel particulates have sizes of significantly less than 1 µm. As such, they represent a mixture of fine, ultrafine, and nanoparticles (Majewski & Khair 2006: 134.) A typical size distribution of heavy-duty diesel exhaust particulates is shown in Figure 4. Note that a logarithmic scale is used for particle aerodynamic diameter.

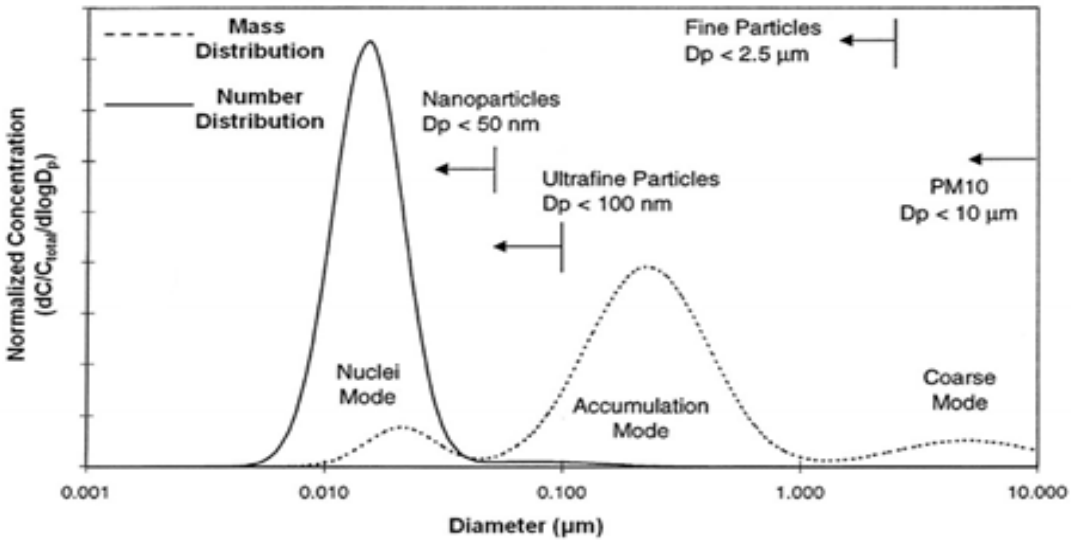

Figure 4. Schematic of mass- and number based concentration of typical particle size distribution from diesel exhaust (Guan & al. 2015).

Size distributions of diesel particulates have a well-established bimodal character that corresponds to the particle nucleation and agglomeration mechanism. In number distributions, most particles are found in the nuclei mode, while most of the particle mass exists in the accumulation mode. (Majewski & Khair 2006: 134.)

Nuclei mode particulates constitute the majority of the particle number, of the order of 90 %, but only a few percent of the PM mass. The nuclei mode typically consists of particles in the 3–30 nm diameter range, which places nuclei mode particles within the nanoparticle range. Nuclei mode particles are primarily volatile and consist mainly of hydrocarbon and hydrated sulphuric acid condensates that are formed from gaseous precursors as the temperature decreases in the exhaust system and after mixing with cold air. Although these particles are volatile they may be relatively insoluble – this could influence their behaviour in biological systems. A small amount of nuclei mode particles may consist of solid material, such as carbon or metallic ash from metals in lube oil and metallic fuel additives. (Kittelson 2006; Majewski & Khair 2006: 135.)

Most of the particle mass – but only a small proportion of the total particle number – exists in the accumulation mode in the diameter range 30–500 nm, so the accumulation mode extends through the fine, ultra-fine and the upper end of the nanoparticle range. Accumulation mode particulates are formed by agglomeration of primary carbon particles and other solid materials, accompanied by adsorption of gases and condensation vapours. They are composed mainly of solid carbon mixed with condensed heavy hydrocarbons, but may also include sulphur compounds, metallic ash, and cylinder wear metals. Accumulation mode particles have been sharply reduced by improved engine technology. (Kittelson 2006; Majewski & Khair 2006: 127-135.)

Coarse mode particles with aerodynamic diameters above 1000 nm contain 5–20 % of the total PM mass and make practically no contribution to particle numbers. The coarse particles are not generated in the diesel combustion process. Coarse mode consists of accumulation mode particles that have been deposited on cylinder and exhaust system surfaces and later re-entrained. (Kittelson 1998; Majewski & Khair 2006: 135.)

It should be noted that the composition of PM might vary widely depending on the engine design, the management strategies, the operating conditions, and the fuel and oil used. (Guan et al. 2015.)

3.3 Effects on Human Health

Numerous epidemiologic studies in recent years have consistently linked PM in the ambient air to negative health effects for exposed populations. Scientific evidence has also shown that ambient particulates of smaller diameters, less than 2.5 µm, are more harmful to humans than coarse particles having diameters of less than 10 µm. This finding was reflected by the introduction of new ambient air quality standards for particles below 2.5 µm by the Environmental Protection Agency (EPA) in 1997. Diesel particulates, with practically all particles being smaller than 1 µm, are entirely within the $PM_{2.5}$ category. (Majewski & Khair: 167.)

The toxicity of particulate matter primarily relates to the number of particles encountered, as well as their size, surface area, and chemical composition (Mills et al. 2009). With regard to $PM_{2.5}$, various toxicological and physiological considerations suggest that fine particles play the largest role in affecting human health. They may be more toxic because they include sulphates, nitrates, acids, metals, and particles with various chemicals adsorbed onto their surfaces. (Pope & Dockery 2006.) Relative to larger particles, particles indicated by $PM_{2.5}$ can be breathed more deeply into the lungs, remain suspended for longer periods of time, penetrate more readily into indoor environments, and are transported over much longer distances (Pope & Dockery 2006).

The health effects of inhalable PM are well documented. They are due to exposure over both the short term (hours, days) and long term (months, years) and include, inter alia:

- respiratory and cardiovascular morbidity, such as aggravation of asthma, respiratory symptoms and an increase in hospital admissions

- mortality from cardiovascular and respiratory diseases and from lung cancer. (WHO 2013: 6.)

The primary exposure mechanism to diesel particulate matter (DPM) is via inhalation. Upon inhalation, particles deposit in the human respiratory system in a size-dependent manner. (Ristovski et al. 2012.) Only particles less than 10 µm in diameter can be inhaled deep into the lungs (Mills et al. 2009). Particles in ultra-fine and nanometric ranges can readily gain access even to the alveolar region of the lung (Ristovski et al. 2012).

There is a vast body of epidemiological literature relating increases in ambient PM exposure to a range of respiratory health outcomes. Short-term exposures may cause symptoms of irritation of the airways, coughing, respiratory infections and compromised pulmonary function, and asthma attacks. Repeated exposure to particulates has been associated with asthma illness, lung function decrements, lung cancer and COPD (Chronic Obstructive Pulmonary Disease). (Ristovski et al. 2012.)

Ristovski et al. (2012) researched the physicochemical properties of diesel particulate matter that are relevant from a respiratory health perspective. It was noted that the particle surface area and organic carbon content of DPM plays an important role in DPM toxicity. The particle surface area influences how toxic compounds adsorb or condense upon particles. Adsorbed organic compounds then greatly affect the chemical and cellular processes that can lead to the development of adverse respiratory health effects. A special concern was the fact that the organic fraction of diesel particulate matter is especially complex, containing hundreds or thousands of soluble organic compounds including PAHs. (Ristovski et al. 2012.) PAHs are known carcinogens and are directly toxic to cells. The exhaust from diesel engines is classified by the International Agency for Research on Cancer as carcinogenic to humans. (WHO 2013: 6.)

In addition to the role of organics, several studies have postulated that transition metals, such as iron, nickel, cobalt, copper and chromium, are possible mediators of DPM-induced airway inflammation. These metals are believed to contribute to particle-induced formation of reactive oxygen species (ROS), which may result in significant damage to cell structures. This is known as oxidative stress. (Ristovski et al. 2012.)

Inhaled, insoluble, ultrafine PM or nanoparticles are even able to cross the alveolar-blood barrier and translocate into circulation, with the potential for impacts on cardiovascular integrity. Once in circulation, nanoparticles may interact with the vascular endothelium or have direct effects on atherosclerotic plaques and cause local oxidative stress and pro-inflammatory effects similar to those seen in the lungs. Increased inflammation may destabilize coronary plaques, which might result thrombus formation, and is the major cause of acute coronary syndromes and cardiovascular death. Long-term exposure to particulate air pollution is also linked to an increase in the risk of venous thromboembolic disease. (Mills et al. 2009.)

Short-term exposure to PM is associated with acute coronary events, ventricular arrhythmia, stroke, and hospital admissions and death caused by both heart failure and ischemic heart disease. Long-term exposure to ultrafine particles increases the lifetime

risk of death from coronary heart disease. The main mediator of these adverse health effects seems to be combustion-derived nanoparticles that contain reactive organic and transition metal components. Inhalation of this particulate matter leads to pulmonary inflammation with secondary systemic effects or, after translocation from the lung into circulation, to direct toxic cardiovascular effects. Through the induction of cellular oxidative stress, particulate matter increases the development and progression of atherosclerosis in the carotid and coronary blood vessels. (Mills et al. 2009.)

Figure 5 provides a schema of mechanistic pathways linking particulate matter with cardiopulmonary disease.

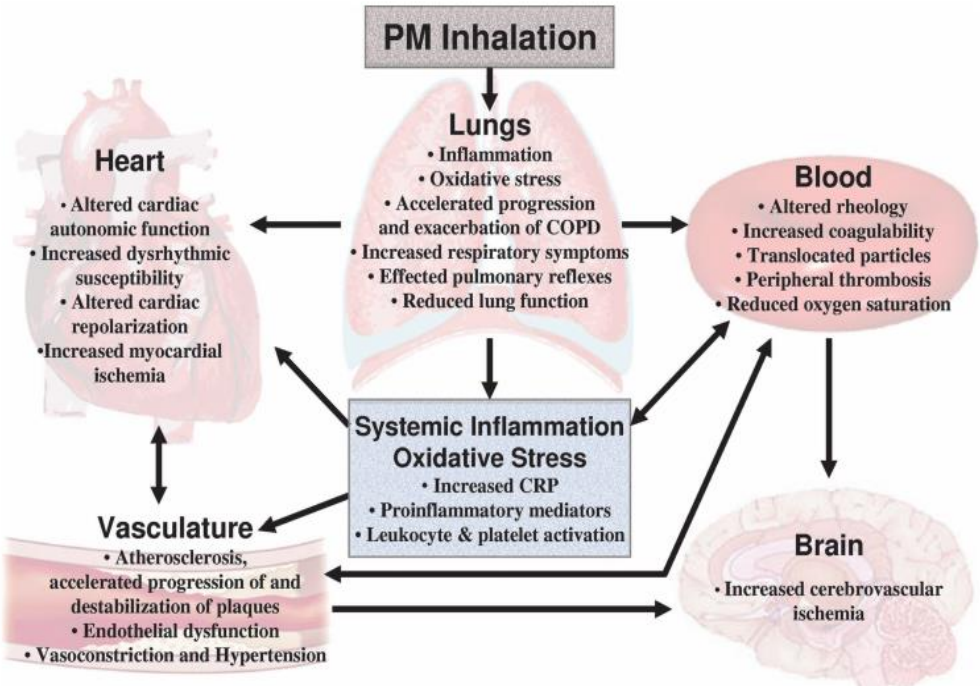

Figure 5. Potential general pathophysiological pathways linking PM exposure with cardiopulmonary morbidity and mortality (Pope & Dockery 2006).

While the primary route by which DPM causes health effects is via inhalation through the human respiratory system, other particle exposure pathways are possible. Translocation is a route of exposure, whereby particles can migrate to a secondary organ, for example the brain, after inhalation, thereby causing health effect in that

secondary organ. (Ristovski et al. 2012.) Recently, there have been increasing reports indicating that inhaled nanoparticles may be associated with neurodegeneration (Win-Shwe & Fujimaki 2011).

Nanoparticles deposited in the nasal mucosa may enter the brain via the olfactory bulb. Another portal of entry of nanoparticles to the brain is from systemic circulation. In the brain, nanoparticles may induce inflammation, apoptosis (cell death) and oxidative stress, which have been experimentally implicated in the pathogenesis of neurodegenerative disorders, such as Alzheimer's disease and Parkinson's disease, and primary brain tumours. (Win-Shwe & Fujimaki 2011.)

There is no evidence of a safe level of exposure or a threshold below which no adverse health effects occur (WHO 2013: 12). In view of the increasing evidence on the adverse health effects of particulate matter and worldwide air quality problems, future diesel engines will have to adopt measures for much more effective control of diesel particulate matter (Majewski & Khair: 150).

4 EU REGULATIONS FOR INLAND WATERWAY TRANSPORT

4.1 Emission standards

The first European Union -wide obligatory emission limits for non-road mobile machinery (NRMM), Directive 97/68/EC, became effective in January 1999. However, engines of inland waterway vessels were not regulated until April 2004 under Amending Directive 2004/26/EC. This directive defines standards Stage III A to cover engines from 19 to 560 kW including constant speed engines, railcars, locomotives and inland waterway vessels. (EU Directive 97/68/EC; EU Directive 2004/26/EC; Transportpolicy 2015.)

In the currently applicable Stage III A standards, engines used in IWT are divided into categories based on the swept volume per cylinder and net power output. The engine categories and the standards are harmonized with the US standards for marine engines. The regulations, given for CO (carbon monoxide), HC + NO_X (hydrocarbons + nitrogen oxides) and PM, can be found in Table 1. The permitted emissions are expressed as mass of emissions per engine work (g/kWh) and depend on the cylinder volume and the net power of the engine.

Table 1. Stage III A emission standards for inland waterway vessels (DieselNet 2015a).

Category	Displacement (D)	Date	CO	HC+NOx	PM
	dm^3 per cylinder		*g/kWh*		
V1:1	D ≤ 0.9, P > 37 kW	2007	5.0	7.5	0.40
V1:2	0.9 < D ≤ 1.2		5.0	7.2	0.30
V1:3	1.2 < D ≤ 2.5		5.0	7.2	0.20
V1:4	2.5 < D ≤ 5	2009	5.0	7.2	0.20
V2:1	5 < D ≤ 15		5.0	7.8	0.27
V2:2	15 < D ≤ 20, P ≤ 3300 kW		5.0	8.7	0.50
V2:3	15 < D ≤ 20, P > 3300 kW		5.0	9.8	0.50
V2:4	20 < D ≤ 25		5.0	9.8	0.50
V2:5	25 < D ≤ 30		5.0	11.0	0.50

Compliance is determined by running the engine at a standardised test cycle. In the European Union, the standard used for emission certification and type approval testing is ISO 8178. ISO 8178 includes a collection of steady-state engine dynamometer test cycles designed for different classes of engines and equipment. The ISO 8178 test cycle – or its 8-mode schedule C1 in particular – is also referred to as the Non-Road Steady Cycle, NRSC. (DieselNet 2015b.)

To represent emissions during real conditions, a new transient test procedure – the Non-Road Transient Cycle (NRTC) – was developed by the US EPA in cooperation with the authorities in the EU. The NRTC is run twice – with a cold start and a hot start. The final emission results are weighted averages of 10 % for the cold start and 90 % for the hot start run. The NRTC will be used in parallel with the prior steady-state schedule NRSC. (DieselNet 2015a.)

For Stage III A, the NRSC (steady-state) is used. The NRTC (transient) can be used for Stage III A testing by the choice of the manufacturer (DieselNet 2015a).

However, despite Directive 97/68/EC being amended a number of times, several technical reviews have concluded that the legislation in its current form has shortcomings. New emission stages were last introduced in 2004 and no longer reflect the current state of technology. The scope of the directive is also overly restricted as it leaves out some engine categories. Furthermore, there is recent conclusive evidence on the injurious health effects of diesel exhaust emissions, especially about PM. One of the main findings is that the size of particles is a crucial factor behind the observed health effects. This point can only be addressed by limit values that are based on the particulate number (PN) in addition to particle mass (PM). (EU Proposal 2014/0581.)

Consequently, on 25th September 2014, the European Commission proposed a new regulation on requirements relating to emission limits and type-approval for internal combustion engines for non-road mobile machinery, COM/2014/0581. This proposal includes the introduction of a new emission stage (Stage V). The stage includes a number of important changes. It widens the scope of regulated engines, including

engines above 37 kW used for the propulsion of inland waterway vessels and auxiliary engines above 560 kW used in inland waterway vessels. Stage V also tightens emission limits for some engine categories, including engines for inland navigation. Furthermore, Stage V adopts particle number emissions limits for several categories. (DieselNet 2015a.)

The new regulation will be applied from 1st January 2017 (EU Proposal 2014/0581). From Stage V, a new regulation will specify emission requirements for all categories of compression ignition and spark-ignition mobile non-road engines, replacing Directive 97/68/EC and its amendments (DieselNet 2015a). Stage V standards, shown in Table 2, would be applicable to propulsion engines above 37 kW and to auxiliary engines above 560 kW, including engines of all types of ignition.

Table 2. Proposed Stage V emission standards for inland waterway vessels (DieselNet 2015a).

Category	Net Power	Date	CO	HC[a]	NOx	PM	PN
	kW		*g/kWh*				*1/kWh*
Propulsion Engines—Category IWP							
IWP-v/c-1	37 ≤P < 75	2019	5.00	4.70[b]		0.30[b]	-
IWP-v/c-2	75 ≤ P < 130	2019	5.00	5.40[b]		0.14	-
IWP-v/c-3	130 ≤ P < 300	2019	3.50	1.00	2.10	0.11	-
IWP-v/c-4	300 ≤ P < 1000	2020	3.50	0.19	1.20	0.02	1×10^{12}
IWP-v/c-5	P ≥ 1000	2021	3.50	0.19	0.40	0.01	1×10^{12}
Auxiliary Engines—Category IWA							
IWA-v/c-1	560 ≤ P < 1000	2020	3.50	0.19	1.20	0.02	1×10^{12}
IWA-v/c-2	P ≥ 1000	2021	3.50	0.19	0.40	0.01	1×10^{12}

[a] A = 6.00 for gas engines
[b] HC + NOx

Auxiliary engines below 560 kW should meet emission standards for category NRE as presented in Table 3 (DieselNet 2015a).

Table 3. Proposed Stage V emission standards for category NRE (DieselNet 2015a).

Category	Ign.	Net Power	Date	CO	HC	NOx	PM	PN
		kW		*g/kWh*				*1/kWh*
NRE-v/c-1	CI	P < 8	2019	8.00	7.50[a,c]		0.40[b]	-
NRE-v/c-2	CI	8 ≤ P < 19	2019	6.60	7.50[a,c]		0.40	-
NRE-v/c-3	CI	19 ≤ P < 37	2019	5.00	4.70[a,c]		0.015	1×10^{12}
NRE-v/c-4	CI	37 ≤ P < 56	2019	5.00	4.70[a,c]		0.015	1×10^{12}
NRE-v/c-5	All	56 ≤ P < 130	2020	5.00	0.19[c]	0.40	0.015	1×10^{12}
NRE-v/c-6	All	130 ≤ P ≤ 560	2019	3.50	0.19[c]	0.40	0.015	1×10^{12}
NRE-v/c-7	All	P > 560	2019	3.50	0.19[d]	3.50	0.045	-

[a] HC+NOx
[b] 0.60 for hand-startable, air-cooled direct injection engines
[c] A = 1.10 for gas engines
[d] A = 6.00 for gas engines

Note that the dates listed in Table 2 and Table 3 are the market placement dates (first registration dates). In most cases, new type approval dates, after which all new engines placed on the market must meet the standards, are one year before the respective market placement dates. (DieselNet 2015a.)

The new regulation is expected to relieve the pressure on member states to take additional regulatory action that could complicate the internal market. The proposal also seeks to remove obstacles to external trade through harmonised rules, in particular with a view to bringing EU and US requirements closer together. Furthermore, the aim of the proposal is to contribute the competitiveness of European industry by simplifying the existing type-approval legislation, improving transparency and alleviating administrative burden. (EU Proposal 2014/0581.)

4.2 Sulphur Content of the Fuel

The quality of fuel used in inland navigation is governed by EU Directive 2009/30/EC. The directive defines, amongst other things, the maximum sulphur content of the fuel used in inland waterway transport. Fuel sulphur content used by inland navigation has been limited in the European Union since 2008 when a limit of 1000 ppm-wt. = 0.1 % sulphur (S) was set. From the beginning of 2011, the maximum allowed sulphur content in fuel is 10 ppm-wt. (10 mg/kg) and therefore has the same maximum sulphur content as fuel for road transport, resulting in a strong reduction of the emission of SO_2. Hence, SO_2 emissions are no longer a real issue for inland waterway transport. (EU Directive 2009/30/EC 2009; Panteia 2013.)

4.3 Abatement Measures for Diesel Particulates

Abatement measures for diesel particulates can be divided into three categories: improvement of fuel and lube oil quality, improvement of the engine combustion process, and exhaust gas cleaning. Typically, 50–70 % of the particulate composition comprises compounds that are related directly to the quality of the fuel, notably its sulphur and ash contents, and cannot be reduced by improved combustion. (Woodyard 2009: 82.)

In the past, it was initially possible to lower exhaust gas emissions by improving engine combustion with corresponding decreases of raw emissions. Although raw emissions are continuing to decrease, this will no longer suffice in the future taking into consideration the developments in the emission legislation (Tschöke et al. 2010: 455.) Even significant improvement in engine combustion will not necessarily result in an adequate reduction of particulate emissions (Woodyard 2009: 82). Meeting these standards will eventually require inland waterway vessels to apply exhaust gas after-treatment technologies.

The term exhaust gas after-treatment includes the systems located in the exhaust gas system with the primary function of reducing engine emissions: catalytic converters, particulate filters, and auxiliary systems that may introduce a reductant or support particulate filter regeneration (Tschöke et al. 2010: 455–456). These technologies are described in the next chapters (5–7).

5 DIESEL OXIDATION CATALYST

The primary function of the diesel oxidation catalyst (DOC) is to oxidize engine carbon monoxide (CO) and hydrocarbon (HC) emissions with the residual oxygen of the exhaust gas into H_2O and CO_2. In addition, DOC exhibits a high activity in the oxidation of the soluble organic fraction of particulate matter, thus reducing total particulate emissions. (Tschöke et al. 2010: 457.) Furthermore, additional benefits of the DOC include oxidation of several non-regulated, hydrocarbon-derived emissions, such as aldehydes and PAHs. Diesel oxidation catalysts may also be effective in controlling diesel odour. (Majewski & Khair 2006: 404–407.)

The converter body consists of a ceramic or metallic honeycomb structure in which the exhaust gas is routed through many small parallel channels as shown in Figure 6. The honeycomb structure enables a high catalytic contact area to exhaust gases. The inner channel walls are covered with an active catalytic coating containing catalytically active precious metal, generally platinum group metal (platinum or palladium). Exhaust gas components, CO and hydrocarbons, diffuse onto this coat when they flow through the converter body and are oxidized (Tschöke et al. 2010: 457.) Reaction products, CO_2 and water vapour, desorb from the catalytic site and diffuse to the bulk of exhaust gas, as shown in Figure 7. (Majewski & Khair 2006: 408).

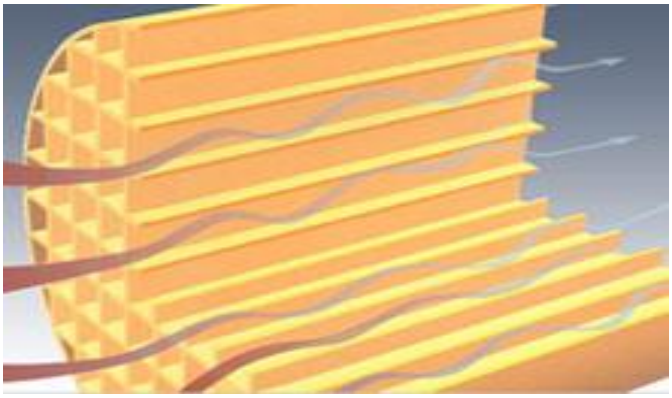

Figure 6. Honeycomb monolith structure (Logical 2015).

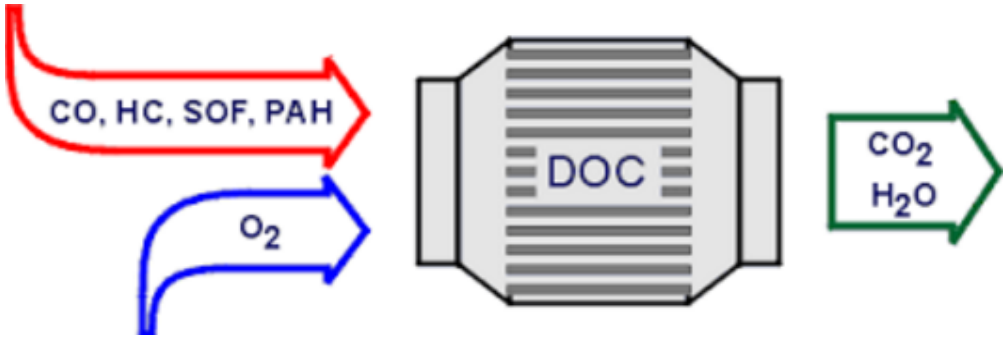

Figure 7. Conversion of pollutants in diesel oxidation catalyst (Majewski & Khair 2006: 404).

The emission reduction is based on the following chemical reactions (Majewski 2015a):

$$HC + O_2 \rightarrow CO_2 + H_2O \quad (2)$$

$$CO + \tfrac{1}{2} O_2 \rightarrow CO_2 \quad (3)$$

Hydrocarbons are oxidized to form carbon dioxide and water vapour. In fact, reaction (2) represents two processes; the oxidation of gas phase HC, as well as the oxidation of SOF compounds. Reaction (3) describes the oxidation of carbon monoxide to carbon dioxide. Since carbon dioxide and water vapour are considered harmless, the above reactions bring an obvious emission benefit. (Majewski 2015a.)

However, an oxidation catalyst will promote oxidation of all compounds of a reducing character, and some of the oxidation reactions can produce undesirable products. The sulphur content of diesel fuel is critical to DOC applications. There is a risk that the DOCs will promote the oxidation of sulphur dioxide (SO_2) to sulphur trioxide (SO_3) with the subsequent generation of sulphate particulates, and actually increase the total PM emissions despite the decrease of the SOF fraction. (Majewski & Khair 2006: 407; Guan et al. 2015.) Hence, usage of ultra-low-sulphur fuel is a precondition for the application of oxidation catalyst.

Additionally, the DOC forms NO_2 due to the oxidation of NO:

$$NO + \tfrac{1}{2} O_2 \rightarrow NO_2 \quad (4)$$

This step is conductive to nitrogen oxide reduction, especially for the selective catalytic reduction (SCR) process. Nitrogen dioxide can also be effectively used for passive soot filter regeneration. (Tschöke et al. 2010: 457–459.) NO_2 regeneration is discussed in more detail in Section 6.3.

The diesel oxidation catalyst can also be used as a catalytic burner. DOC releases reaction heat when CO and HC are oxidised. This increases the temperature of the exhaust gas system after DOC, and is applied to facilitate the increase in temperature necessary for particulate filter regeneration. (Tschöke et al. 2010: 457.)

The level of particulate matter reduction is influenced in part by the percentage of soluble organic fraction in the particulate. According to Papadimitriou et al. (2015: 52), DOC exhibits the total PM reduction efficiency of 20–40 %. The PM removed by DOCs is mainly soluble organic fraction from unburned fuel and oil. DOCs generally have insignificant impact on elemental carbon (EPA 2010). DOCs also reduce HC emissions (40–70 %) and CO emissions (40–60 %) (Papadimitriou et al. 2015: 52). Majewski & Khair (2006: 404) evaluate HC and CO emissions reduction efficiencies as up to 90 % at sufficiently high exhaust temperatures.

DOC has low installation costs, is easy to install, and little or no maintenance is required (Papadimitriou et al. 2015: 52). However, two catalyst deterioration mechanisms are fundamental. First, the precious metal particulates can agglomerate at very high exhaust gas temperatures. This reduces the specific surface area of the precious metals. Second, catalyst poisons can coat the precious metal surface and inhibit their accessibility. The best known catalyst poison is the sulphur contained in the fuel. (Tschöke et al 2010: 458.) However, in inland navigation in the EU, sulphur poisoning of DOC is no longer considered a problem due to the ultra-low-sulphur fuel.

Diesel oxidation catalysts can be coupled with SCR or lean NO_x catalysts for additional reductions. They can also be integrated with diesel particulate filters. DOCs have also been shown to be effective with biodiesel and emulsified diesel fuels, diesel blends and other alternative diesel fuels. (Papadimitriou et al. 2015: 52.)

6 DIESEL PARTICULATE FILTER

In order to remove insoluble particulate matter – elemental carbon and ash – from the exhaust gas, diesel particulate filters (DPF) are necessary (Merkisz & Pielecha 2015: 109). The function of DPFs is to physically capture a very large fraction of diesel particulates from the exhaust gas flow, thus preventing their release into the atmosphere (Majewski & Khair 2006: 459; Tschöke et al. 2010: 458).

Diesel particulate filters capture particulate emissions through a combination of filtration mechanisms: diffusional deposition, inertial deposition and flow-line interception (Majewski 2015b), see Figure 8. Due to the overlapping filtering mechanisms, both large as well as small soot particles can be held back, thus achieving extremely high filtering efficiency across the entire spectrum of sizes. Since nearly all emitted particles are smaller than the pores of filter substrate, they are not caught in the filter due to their size but mostly by means of diffusion. While the diffusion speed increases with decreasing particulate size, smaller particulates are actually separated the most effectively. With rising soot loads, there is a transition from depth filtration in the filter wall down to surface-type filtration; both the soot layer stored in the pores as well as the soot cake on the filter wall itself act as an effective filtering medium. (Fiebig et al. 2014.)

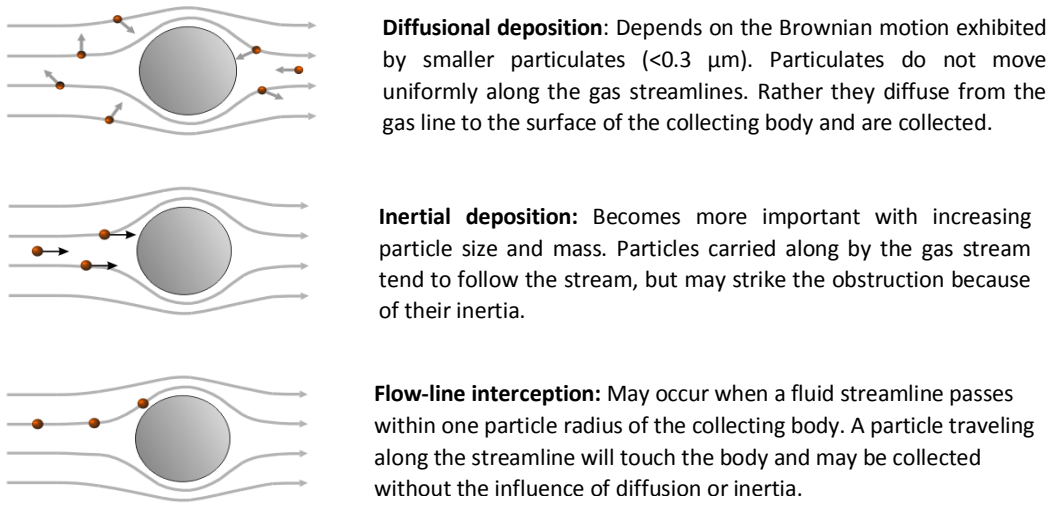

Figure 8. Filtration mechanisms of diesel particulate filters (Majewski 2015b).

6.1 Filter Designs

The substrate of the filtering systems can be made of many different materials and have different compositions. The most common design for a diesel particulate filter is the wall-flow monolith. A number of other, alternative filter designs and materials have also been developed. Examples include cartridges made of ceramic fibres, various types of ceramic foams, and metallic materials, such as metal fibre felts and sintered metal structures. (Majewski & Khair 2006: 460).

Ceramic wall-flow monoliths are by far the most common type of DPF. The wall-flow filter consists of several small parallel channels running axially through the part. Adjacent channels are alternatively plugged at each end in order to force the diesel exhaust gas flow through the porous substrate walls between the channels. These walls act as a mechanical filter, see Figure 9. (Majewski & Khair 2006: 460.)

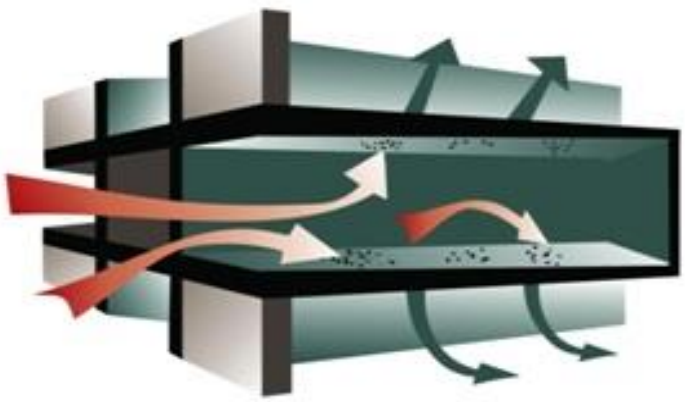

Figure 9. Wall-flow monolith structure (Nett Technologies Inc. 2015).

Wall-flow monoliths are characterized by high filtration efficiencies from 70 to 95 % of total PM. Even higher efficiencies (>99 %) are observed for solid particulate matter fractions. (Konstandopoulos 2013: 25; Majewski 2015e.)

Drawbacks of monolith filters include a relatively high pressure drop. As the solid fraction of PM (soot) accumulates on the channel walls, the pressure drop across the

filter increases, leading to increased back pressure to the engine and deteriorated fuel economy. Hence, an active regeneration of the filter by removal of the soot is a relevant part of wall-flow operation. (Konstandopoulos 2013: 8.) Regeneration methods are discussed in more detail in Section 6.3.

In addition, increased maintenance concerning wall-flow filters is related to ash emissions. As the incombustible ash particles cannot be removed from the filter through thermal regeneration, the accumulated ash must be physically removed from the DPF. This generally involves ash cleaning at a dealer or service centre. The most prevalent method of ash cleaning and removal from diesel particulate filters is reverse flow pneumatic cleaning. The basic cleaning method comprises driving a flow through the filter from the outlet side (reverse flow) and collecting the ash blown out of the filter in a dust collection system. Before the pneumatic cleaning, the filter may be placed in a furnace and heated in order to oxidize any residual soot. Various commercial cleaning systems also exist, which utilize either the localised application of high pressure air, or low pressure but high volume flow through the entire filter cross-section. Ash is classified as a hazardous waste and must be handled and disposed of as such. (Sappok 2016.)

For reducing the blocking risk of the filter and to avoid complex regeneration and cleaning procedures, an alternative approach has been developed for soot particle removal: an open particulate filter. The open particulate filter, also called a flow-through or partial flow filter, or particle oxidation catalyst (POC), has no risk of clogging because it employs a structure with open channels instead of alternately plugged channels with porous walls (Feng et al. 2014). Open flow-through passages allow exhaust gases to flow, even if the soot loading capacity of the filter is saturated. Another main advantage of this method is the lower exhaust back pressure compared to DPF.

The structure of the POC is similar to a diesel oxidation catalyst. In addition, it utilises substrate with some capacity to capture solid particles. The PM reduction efficiency of POC is higher than that of the DOC, but lower than DPFs, see Figure 10.

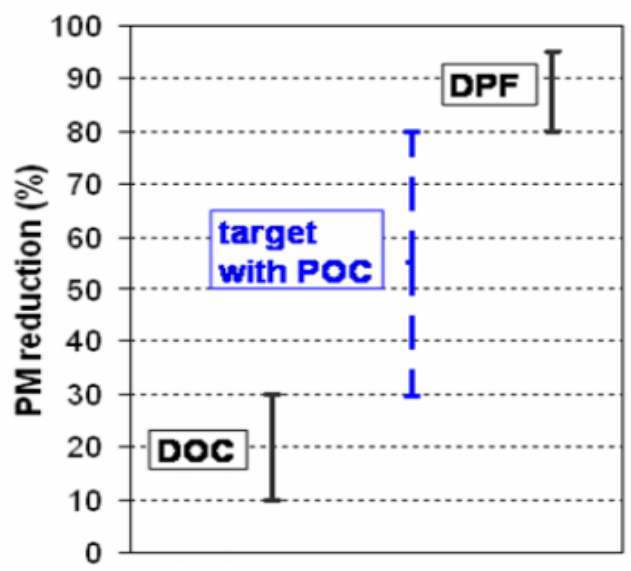

Figure 10. PM reduction target of POC (Kinnunen 2009).

The filter element can be made of a variety of materials and designs. Flow-through filters employ metal wire mesh structures or tortuous flow, metal foil-based substrates with metal fleece or sintered metal sheets. In the design illustrated in Figure 11, structural elements are used to divert a part of the exhaust gas flow through a fibre fleece and into the filter's adjacent channels, filtering out particulate matter. (MECA 2007: 14.)

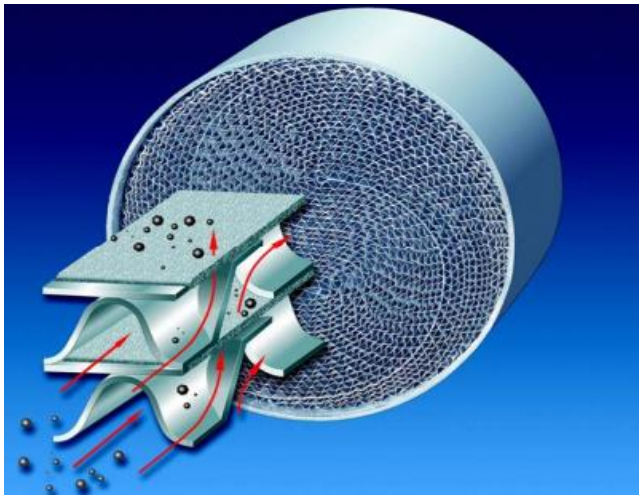

Figure 11. Metallic flow-through filter made up of corrugated metal foil and layers of porous metal fleece (MECA 2007: 15).

To maintain the filtration efficiency of POC, the removal of trapped particles is needed. Most POCs rely on passive regeneration, i.e. soot is removed through catalytic reactions. (Guan et al. 2015.) The filter can be coated with catalyst materials to assist in oxidizing the soot, or the filter can be used in conjunction with an upstream diesel oxidation catalyst (MECA 2007: 14).

The use of ultra-low-sulphur fuel is a prerequisite for the effectiveness of a POC system. High sulphur contents would lead to the generation of sulphate particles and might dramatically increase PM. With ULSD, POCs may provide 30–85 % reduction of PM mass from diesel exhaust. POC systems with catalyst coating or upstream DOC offer also the co-benefits of reducing CO, HC and toxics by up to 80–90 %. However, the reduction efficiency is significantly lower than that of wall-flow DPF. (Guan et al. 2015.)

An advantage of this technology is that flow-through filters generally do not accumulate inorganic ash constituents present in diesel exhaust, and usually do not require ash cleaning. (MECA 2007: 14.) In addition, as already mentioned, POC is much less likely to plug. As such, POC is considered a low-cost and safe PM reduction technology. However, plugging may occur under low-load conditions, and therefore active regeneration may be needed. Furthermore, “blow-off” issues may occur due to the open channels of the POC. Particulates which were already deposited on a filter may be re-entrained by the gas, and superfluous particles escape into the atmosphere, causing the observed PM filtration efficiency to decrease and even to become negative. (Feng et al. 2014; Guan et al. 2015.)

6.2 Filter Materials

The filter substrate is a key component of the diesel filter system, affecting both its performance and durability. Design targets for DPF materials include high filtration efficiency and low pressure drop, high maximum use temperature, high soot holding

capacity, high thermal shock resistance, chemical stability and compatibility with regeneration methods. (Majewski & Khair 2006: 473.)

A list of the most common filter materials includes extruded ceramic materials, ceramic fibres and metallic materials, such as metal fibres and sintered metal powders. An estimated distribution of these materials on the global particulate filter market can be seen in Table 4.

Table 4. Distribution of DPF materials for exhaust cleaning (Konstandopoulos 2013:7).

Material	Utilization
Extruded ceramics	70%
Ceramic fibres (wound and knitted)	25%
Sintered metals (granular or fibrous)	4%
All others	1%

Extruded ceramic materials are typically used in wall-flow monoliths. Two materials most commonly used in commercially available monoliths are cordierite and silicon carbide (SiC) (Konstandopoulos 2013: 25.) Cordierite is characterized by a very low thermal expansion coefficient, which makes it resistant to extreme thermal cycling. It also exhibits high temperature resistance and good mechanical strength. The drawback of cordierite is the low melting point of 1450˚C, which has been considered insufficient under certain uncontrolled regeneration conditions. Silicon carbide is characterized by a higher operating limit (1800–2400˚C) and favourable pore network structure. The drawback of SiC is a higher thermal expansion coefficient and higher cost. A comparison of cordierite and silicon carbide materials is shown in Table 5. (Majewski & Khair 2006: 474-485.)

The newest commercial filter monolith material is aluminum titanate (AT). Aluminum titanate is more generally being considered for heavy duty applications due to its excellent thermal shock resistance, allowing single piece monoliths to be used even for larger size DPFs. (Konstandopoulos 2013: 26.)

Table 5. Comparison of cordierite and silicon carbide materials (Majewski & Khair 2006; 482).

Property	Cordierite	SiC	Comment
Melting temperature (°C)	1450	2400	Cordierite melting temperature, combined with its low heat capacity, is insufficient to protect the material during uncontrolled regenerations.
Thermal expansion, CTE (1/°C)	0.7×10^{-6}	4.5×10^{-6}	Cordierite has a superior thermal shock resistance (low thermal expansion).
Thermal shock parameter, TSP	790	124	
Permeability ($\times10^{-12}$ m^2)	0.50	1.24	SiC has uniform network of open pores resulting in higher permeability and lower ΔP.
Volumetric heat capacity (kJ/m^3K)	500	950	Higher heat capacity and conductivity in SiC are believed to be unfavourable for sustaining regeneration, but provide greater margin of safety in uncontrolled regeneration.
Thermal conductivity (W/mK)	<2	20	
Relative cost	Low	High	SiC requires more expensive raw materials and is processed at higher temperatures.

Different types of high-temperature ceramic fibres have been used in numerous designs of diesel filtration media. Advantages of ceramic fibres include their very good thermal stability, tolerance to ash particle accumulation and high noise attenuation properties (Majewski & Khair 2006: 474, 491). In contrast, the main disadvantage of fibre filters is their incompatibility with catalyst coating. Suitable regeneration methods for fibre filters include fuel additives, fuel burners, and electric heaters. Ceramic fibre filters also appear to have somewhat lower filtration efficiency than monolithic filters. Moreover, they tend to occupy more space in the exhaust system (larger size compared to monolithic filters) and may emit secondary emissions in case of fibre breakage. Finally,

they do not present any cost advantages compared to ceramic wall-flow filters. (Konstandopoulos 2013: 28–29.)

Porous materials made of high temperature resistant metals are presented as alternatives to ceramics. The most common design is based on a stainless steel wire mesh, which is coated with sintered metal powder mixed with additives (Majewski & Khair 2006: 476). Another option is to use sintered metal fibres (metal fleece) as a filter medium. Wall structures with sintered metal powder on a wire mesh have demonstrated 80–90 % filtration efficiency. Structures involving metal fibres usually come with somewhat lower initial filtration efficiency during the clean state of the filter. After soot has been trapped by the filter material, filtration efficiency increases above 99 %. An additional advantage of fibres is their low thermal mass (faster heat-up). (Konstandopoulos 2013: 27–28.)

The main advantages of sintered metal filters are their lower exhaust backpressure (due to the high porosity of the filter media), mechanical durability and resistance to thermal shock. In addition, with electrically conductive materials, the regeneration can be performed by applying electricity to the filter media. The sintered metal material also offers flexibility in shaping, which can be utilized for the design of DPF systems with higher ash tolerance, and they can be flexibly fabricated in a variety of shapes to accommodate space constraints. On the negative side, they are characterized by their relatively heavy weight. The construction of metal filters is also, on average, more expensive than that of ceramic wall-flow monoliths. (Majewski & Khair 2006: 476; Konstandopoulos 2013: 27–28.)

6.3 Filter Regeneration

All filter materials are designed to hold a certain quantity of soot. If the filter becomes overloaded, the particulates create an obstruction to the gas flow and eventually result in excessively high exhaust gas pressure drop across the filter. This may lead to clogging of the filter itself and can negatively affect the engine operation. Therefore, the DPF

system has to provide a reliable way of removing collected soot from the filter to restore its filtration capacity and to ensure problem-free operation. (Majewski & Khair 2006: 462; Guan et al. 2015.) This removal of accumulated soot particles – known as filter regeneration – can be performed either continuously during the regular operation of the filter or periodically after a pre-determined quantity of soot has been accumulated (Guan et al. 2015).

The oxidation of carbon in diesel soot can occur via reactions with the oxidizing agent O_2 and NO_2 (Guan et al. 2015). The oxidation by O_2 can be described by the following reactions:

$$C + O_2 \rightarrow CO_2 \quad (5)$$

$$2C + O_2 \rightarrow 2CO \quad (6)$$

It should be noted that CO_2 is the preferred product. Increased CO emission may occur if O_2 is depleted in the reaction zone. (Guan et al. 2015.)

O_2 is present in diesel exhaust gas at sufficient concentrations practically in all operating conditions. However, relatively high temperatures (600°C) are required to achieve appreciable regeneration rates with O_2. Thus, the O_2-based regeneration is mostly seen in active DPF systems. (Guan et al. 2015.)

The NO_2-based regeneration occurs via following reactions (Guan et al. 2015):

$$2NO + O_2 \rightarrow 2NO_2 \quad (7)$$

$$C + 2NO_2 \rightarrow CO_2 + 2NO \quad (8)$$

$$C + NO_2 \rightarrow CO + NO \quad (9)$$

NO_2 contains a much more active oxygen species for soot combustions than the O_2 molecule itself, allowing filter regeneration at remarkably lower temperatures; regeneration can be triggered at the temperature as low as 260°C (Guan et al. 2015). NO_2 is present in the native diesel exhaust gas in very small quantities, but the NO_2

concentrations can be increased by using a DOC or other methods. (Majewski & Khair 2006: 471.)

The temperature of the exhaust gas is too low to sustain auto-regeneration of the trap. This problem can be solved by 1) increasing the temperature of the trapped soot to a point where it starts oxidizing, 2) decreasing the required soot combustion temperature to a level that is reached during regular engine operation, or 3) by utilizing NO_2 as a low-temperature agent. The first approach is used in active filter systems, the last two in passive (catalytic) filter systems. (Majewski & Khair 2006: 497.)

Diesel particulate filter systems are designed by combining different filter materials with selected regeneration methods. A classification of DPF systems based on the principle of regeneration is shown in the figure below (Figure 12).

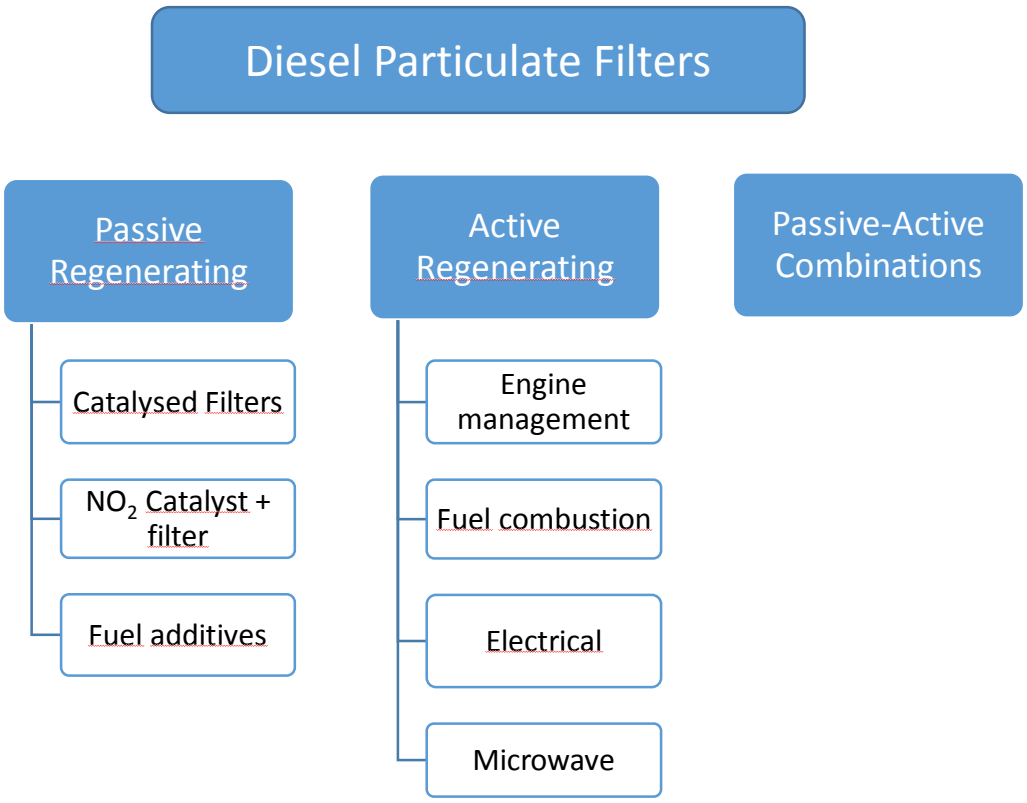

Figure 12. Classification of filter systems by regeneration method.

In passive systems no external energy is applied for the regeneration process. In these systems the soot oxidation temperature is lowered to a level allowing auto-regeneration during regular engine operation. This can be achieved by introducing an oxidation catalyst to the system, either directly onto the filter media surface or added to the diesel fuel as a fuel additive. Passive systems that utilize NO_2 as a low-temperature soot oxidation agent have also been introduced. (Johansen 2015; Majewski 2015c.)

6.3.1 Catalysed Diesel Particulate Filter

In a catalysed diesel particulate filter (CDPF), a catalytic material is coated onto the surface of the filter to lower the ignition temperature necessary for oxidizing accumulated soot. Typically, the CDPF is coated with a catalyst combination of base metal catalysts, such as stabilized cerium oxide, to decrease the temperature of soot oxygen combustion by direct contact with soot, and platinum containing catalyst for generating new NO_2 and for cleaning up CO and HC. Mixed metal oxides with perovskite- and spinel-type structures have also been tested for soot oxidation. (Johansen 2015.)

Most CDPFs use wall-flow monoliths for capturing solid particulates. The efficiency for solid particulates is between 95 % and nearly 100 %. The total filtration efficiency varies between 60 % and 95 %, because the sulphate and SOF transformation depends entirely on chemical reactions that occur on contact with the filter catalyst. CDPFs are also very effective in reducing particle number emissions, provided the nanoparticles are solid. If liquid nanoparticles are created through condensation downstream of the filter, they obviously cannot be controlled by the CDPF. (Majewski & Khair 2006: 502–504.)

Catalysed particulate filters also facilitate a number of oxidation reactions in the gas phase emissions CO and HC, similar to those occurring in the diesel oxidation catalyst. Platinum-based CDPF typically achieves 80–90 % conversion of CO and 60–70 % conversion of hydrocarbons. (DECSE 2000.)

Catalysed filters require an exhaust temperature of about 325–420℃ for regeneration, depending on engine technology and fuel quality (Majewski & Khair 2006: 471). This may constitute an inherent problem: if the exhaust gas temperature is below the catalyst temperature required, CDPFs do not regenerate adequately and can plug up, causing excessive backpressure on the engine (Guan et al. 2015). Thus, the CDPF may require some kind of active regeneration support in cases where excessive soot load is detected in the filter (Majewski & Khair 2006: 507).

6.3.2 NO_2 Catalyst + Filter

The rate of soot oxidation can be increased if an oxidant more active than oxygen is introduced to the particulate filter. Nitrogen dioxide can oxidize diesel particulates at much higher rates than oxygen. A filter system that uses NO_2 for regeneration was first introduced by Johnson Matthey under the trade name CRT® (continuously regenerating technology). (Majewski & Khair 2006: 471.)

The CRT system consists of two functional components packaged into one housing: an oxidation catalyst upstream and a ceramic wall-flow diesel filter downstream (Figure 13). The NO_2 necessary for filter regeneration is generated in the DOC from NO present in diesel exhaust. (Majewski & Khair 2006: 508.)

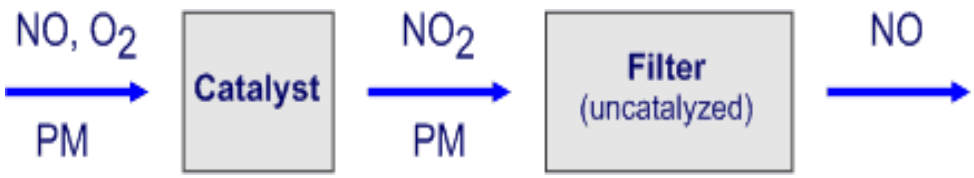

Figure 13. CRT Filter structure and principle of operation (DieselNet 2015c).

The overall PM mass reduction efficiency of CRT depends on the conversion of the three fractions of diesel particulates: solid carbon and ash, SOF, and sulphate particulates. As the CRT utilizes well-proven wall-flow monoliths with more than 95 %

efficiency for solids, and an active DOC that is relatively efficient in oxidizing SOF, the overall PM performance depends on sulphate particulates. A number of studies using 10 to 30 ppm S fuel reported total PM reductions in the range of 90 %. (Majewski & Khair 2006: 512). In addition, the CRT influences the emissions of gaseous pollutants as well as the PM. CO and HC emissions may be reduced by about 85–95 %, aldehydes by 50–90 %, and PAH reduction can be up to 80 %. (Lanni et al. 2001.)

CRT is capable of regenerating at temperatures as low as 250–300°C. According to Johnson Matthey (2015a), the conditions normally required for reliable CRT performance are:

- A significant proportion (~50 %) of the operating time with an exhaust temperature higher than 250°C
- The NO_x/PM ratio in the exhaust gases 20:1 or greater
- The maximum sulphur content of the fuel 50 ppm.

The CRT system has been further refined by combining CRT and CDPF. This system has a DOC upstream from DPF that is also coated with a catalyst. (Guan et al. 2015.) The NO_2 produced by the diesel oxidation catalyst reacts with the PM trapped in the filter, producing NO and CO_2. Some of the NO is then re-oxidised to NO_2 in the filter, which then reacts with more trapped PM. This enables the system to further reduce the regeneration temperature to about 210°C for 40 % of the operating time. CCRT is also able to operate on engines with a low NO_x/PM ratio that is insufficient for a standard CRT system. (Johnson Matthey 2015b.)

One of the side effects in filters which utilize NO_2 regeneration is increased nitrogen dioxide level at the tailpipe, i.e. increased NO_2/NO ratio in the total NO_x emission. This is because the generated NO_2 is never completely consumed in soot oxidation. (Majewski 2015c.)

6.3.3 Filters with Fuel Additives

Decreasing the oxidation temperature of carbon particles can also be achieved by fuel additives, so-called fuel borne catalysts (FBC), directly mixed into the diesel fuel (Guan et al. 2015). Today, iron (Fe) and cerium (Ce) are used as fuel-borne catalysts, and they are added to the diesel fuel as organic compounds at a level of 5–10 ppm (Johansen 2015). The oxidation mechanism is similar to that in catalyst coated traps, but an additional advantage of this method is that the catalyst physically incorporates into the particulates, and not just on the surface of the filter. As a result, the physical contact between soot and the catalyst is much better. (Majewski & Khair 2006: 471.) FBC can be used in conjunction with both passive and active filter systems (Guan et al. 2015).

FBC reduces the DPF regeneration temperature from >600˚C to as low as 350˚C (Guan et al. 2015). The choice of FBC levels depends on the activity of the additive, exhaust gas temperature and composition of PM. In general, higher additive concentrations allow regeneration at lower temperatures. (Majewski & Khair 2006: 515.) However, after the soot oxidizes, the metal oxides from the additive remain in the filter as a residue and, thus, increase the ash fraction that thermal regeneration is unable to remove (Tschöke et al. 2010: 460). Therefore, high levels of additives increase filter pressure drop in short order due to the accumulation of ash. High additive levels also increase operational costs. Other potential disadvantages of FBC are the system's complexity due to automated dosing devices, possible secondary emissions, and impacts on the engine or its components, for example some additives are known to cause fuel injector fouling. (Majewski & Khair 2006: 515.)

A moderate increase in exhaust temperature may be needed in systems utilizing catalysts or fuel additives to lower the soot ignition temperature. Target exhaust temperature in these systems varies between 350 and 450°C. This temperature level can be achieved through several engine management measures, such as post-injection of fuel into the combustion chamber, delayed injection or by increasing the intake air temperature. (Majewski & Khair 2006: 470–471; Merkisz & Pielecha 2015: 117.)

Any application in which the exhaust gas temperature cannot be guaranteed to be suitable for passive regeneration will require some active regeneration system. Active filter regeneration techniques operate by raising the temperature of the soot trapped in the filter to around 600°C, at which the soot will combust in oxygen. To achieve this, it is necessary to increase the exhaust gas temperature, or to increase the temperature of the filter itself. For this, an outside energy source is needed. Hence, the active regeneration process of the filter will lead to increased fuel consumption. However, because active regeneration occurs periodically, the overall fuel penalty is approximately 2%. (Guan et al. 2015.)

6.3.4 Engine Management

The exhaust gas temperature can be increased by the combustion of an additional quantity of diesel fuel, either through in-cylinder engine management methods, or through the injection and combustion of fuel in the exhaust system. In the first approach, the fuel is introduced through late cycle injection (post-injection) using the fuel injection system of the engine. (Guan et al. 2015.) However, all of the post-injected fuel cannot be combusted in the cylinder, resulting in increased HC concentrations in the flue gas. Therefore, in-engine strategies usually require a catalyst to oxidize HCs and further increase the exhaust temperature. (Majewski 2015d.) In addition, it should be noted that the in-cylinder post-injection may cause lubricant dilution by the unburned fuel since a small amount of fuel enters the engine crankcase. Thus, this method is not advisable for heavy-duty applications with long lubricant oil intervals. (Guan et al. 2015.)

6.3.5 Filters Regenerated by Fuel Combustion

Fuel can also be injected to and combusted in the exhaust system to increase the exhaust gas temperature. The main methods are flame combustion and catalytic combustion. A

combination of the above mentioned systems may also be used in certain cases. (Majewski 2015c.)

In flame combustion, the fuel is injected and combusted in a fuel burner, usually with a dedicated supply of combustion air, from which the flame enters the exhaust system (Guan et al. 2015). Burner systems can be designed for regeneration in any engine operating condition. These so-called full-flow burners are the most common burners used in NRMM applications. The operating principle of full-flow burner filters is shown in Figure 14.

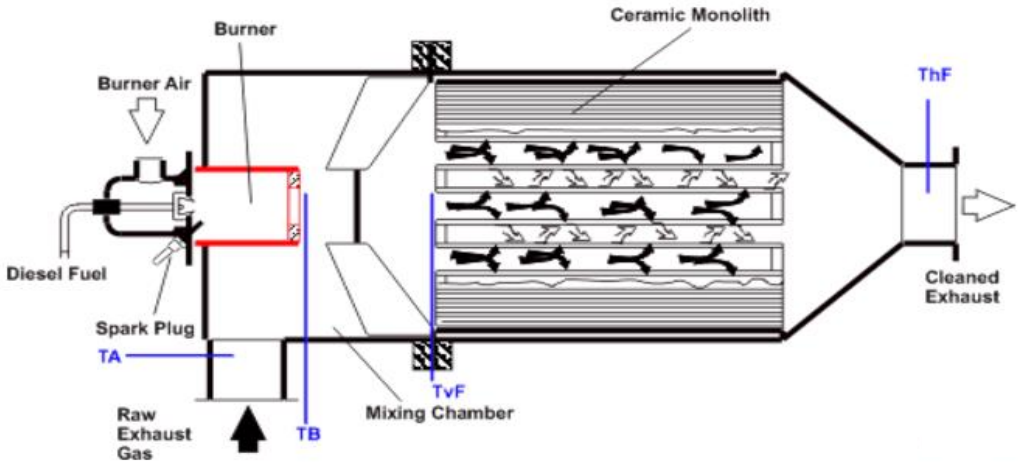

Figure 14. Schematic of full-flow burner filter system (DieselNet 2015d).

The filter unit integrates the ceramic wall-flow monolith, the mixing chamber, and the burner. Versions with ceramic fibre filters have also been developed. Diesel fuel is mixed with air supplied by an electrically driven compressor, injected into the combustion chamber, and ignited by ignition electrodes. The filter control unit starts the burner and controls its operation to maintain the required filter temperature. (Majewski & Khair 2006: 528–529.)

Full-flow fuel burner filters require complex electronic control units to maintain thermally balanced regeneration. The soot load generates quantities of additional heat during regeneration, which may lead to significant increase of the filter temperature and overheating of the unit. (Majewski & Khair 2006: 528.)

In fuel burner systems, the gaseous emissions including NO_x, CO and HC are not affected by the filter. The TPM collection efficiency is about 90 % and the collection efficiency for solid fraction of PM is about 99 %. Reported data indicate that the filter also reduces nanoparticle emissions, as well as a portion of PAH. (Majewski & Khair 2006: 530.)

In catalytic fuel combustion, the fuel is injected into the exhaust gas upstream oxidation catalyst, which is located in front of the DPF. The oxidation of HCs in the DOC increases the exhaust gas temperature to the level needed for regeneration. (Guan et al. 2015.) The catalytic combustion system requires a certain minimum catalyst temperature. Thus, regeneration might not be possible at idle or at light engine load, when the exhaust temperature is low. In the combined burner-catalyst approach, a small burner installed upstream of the catalyst ensures the operation of the catalyst regardless of the engine operating conditions. (Majewski 2015d.)

In comparison to in-cylinder injection, exhaust injection results in a smaller fuel economy penalty, as the heat is released immediately upstream of the filter, eliminating heat losses in the engine. (Majewski 2015d.)

6.3.6 Electrically Regenerated Filters

Electric heating can be used in a number of configurations: the energy can be supplied to the exhaust gas, to the filter substrate, directly to the accumulated soot, or through a combination of these methods. (Majewski & Khair 2006: 521; Guan et al. 2015.)

The most common electric regeneration method involves heating of the exhaust gas, or a stream of air used for regeneration, by resistive heater. This is a relatively simple method of triggering the regeneration of a soot-laden DPF and it can be used with a wide range of filter substrates. In wall-flow monolith filter systems, the electric heater is usually placed upstream of the filter substrate. (Majewski & Khair 2006: 521.) The prime concern related to this method is its high energy consumption, especially if the

entire exhaust gas stream must be heated up (Konstandopoulos 2013: 15). The partial flow layouts and regeneration with hot air stream are considered to be more energy efficient (Guan et al. 2015).

Electrical heating of the filter substrate requires the filter to have an electrically conductive medium, serving as a resistive heater itself. Electrically heated filter substrates include, e.g., sintered metal fibres. If materials of low heat capacity can be used, this method may show good energy efficiency. (Majewski & Khair 2006: 521.)

6.3.7 Microwave Regenerated Filters

Microwave heating is based on absorption of radio frequency energy by dielectric materials. There are two approaches to the microwave-assisted regeneration of DPFs: 1) heating of the soot accumulated in the filter by microwave, and 2) heating both the soot and the microwave-absorbing filter material. The latter approach requires either the filter substrate to be made of radio frequency absorbing material, or a microwave susceptor to be incorporated into the filter substrate or coated onto the substrate surface. (Majewski & Khair 2006: 531–532.)

Microwave regeneration may in principle offer the advantage of fast and selective heating. In addition, there are claims that microwave heating can result in lower soot combustion temperatures than realized by other heating techniques (Palma et al. 2014). Many studies have been performed concerning microwave regenerated filter systems, but there have been very few attempts of fitting this method on a diesel vehicle. However, laboratory experiments have shown a capability of microwave heating to deposit energy directly into the PM accumulated on the filter. Theoretically, this would enable very low energy expenditure due to the minuscule thermal mass of the trapped soot. (Konstandopoulos 2013: 15-16.) Furthermore, by using filter coatings that pair suitable dielectric with catalytic properties, the combined microwave heating with catalytic combustion results in lower soot oxidation temperatures and higher reaction

rates, lowering the time and energy required for the filter regeneration (Palma et al.2014).

The main problems reported with regard to microwave-assisted regeneration are related to the non-homogenous electric field distribution. As a result, the filters tend to show irregular regeneration patterns, incomplete regeneration, or excessive exothermal release leading to substrate damage. (Konstandopoulos 2013: 15.) However, if a satisfactory degree of control over the radio-frequency energy deposition were achieved, this method would offer more flexibility in designing and controlling the regeneration than do the electric and burner approaches (Majewski & Khair 2016: 533).

6.3.8 Passive-Active Combinations

In extended operation conditions under which passive regeneration is not enough to keep the filter clean, active strategies need to be implemented to obtain efficient and reliable regeneration of diesel particulate filters. (Guan et al. 2015.) In passive-active regeneration combinations the catalyst-based filter is also equipped with some kind of active regeneration system (Majewski 2015c).

Passive-active combinations may be able to sustain fully passive operation during periods of increased exhaust temperature at high loads. In the case of idle operation, or continuous light-load conditions with low exhaust gas temperatures when the soot ignition temperature cannot be reached even with catalyst, active regeneration is utilized. (Guan et al. 2015.)

The use of a catalyst allows performing regeneration at a lower temperature and shortening the regeneration time period, compared to non-catalytic active systems. In either case, the fuel economy penalty associated with active regeneration can be minimized. Regeneration at a lower temperature also reduces thermal stress, thus increasing the life span of the filter media. (Majewski 2015c.)

7 ALTERNATIVE TECHNOLOGIES FOR PM EMISSION CONTROL

7.1 Non-thermal Plasma

Non-thermal plasma (NTP) is believed to be a promising candidate for total engine exhaust emission control, including PM and NO_x (Babaei et al. 2014). A plasma, known as the fourth state of matter, is a gas that has been ionised by passing an electrical current through it. There are several ways non-thermal plasma can be generated: dielectric-barrier discharge, corona discharge, electron beam irradiation and microwaves. The basic principle is that electron temperature and gas temperature greatly differ, so that the electron temperature is much higher than the gas temperature. (Manivannan et al. 2014.) High-energy electrons impact with gas molecules and produce various chemically active species, such as oxygen radicals (O), ozone (O_3), hydroxyl radicals (OH) and free nitrogen radicals (N) (Babaei et al. 2014).

Both gaseous and PM emissions of exhaust gases undergo chemical changes when they are exposed to the plasma (Babaei et al 2014). For the removal of elemental carbon, there are two routes by NTP: 1) simple oxidation by means of O and OH radicals, and 2) interaction between NO_2 and carbon soot. Hydrocarbons, including SOF and gaseous HC, initially react with O atoms, and are oxidized into CO and CO_2. (Song et al. 2009.)

Babaei et al. (2014) investigated the effect of non-thermal plasma on particular number and mass concentrations. A dielectric barrier discharge reactor was designed and a sample of diesel exhaust was passed through this reactor to test the effect of NTP. The results showed a considerable reduction of particulate matter both in particulate number and particulate mass. The maximum reduction in PM mass was 72 %, countervailing reduction in PM number was 67 %.

Non-thermal plasma can also be utilized in the regeneration of DPFs. NTP-induced ozone is suggested as a diesel soot oxidant that may enable regeneration at temperatures even lower than those possible with NO_2 (Majewski & Khair 2006: 546).

To confirm the non-thermal plasma DPF regeneration method for marine diesel engines, pilot-scale experiments were performed by Kuwahara et al. (2012). The study confirmed that continuous DPF regeneration can be successfully carried out for a marine diesel engine operated with marine diesel oil containing 0.1 % sulphur. (Kuwahara et al. 2012.)

An important advantage of non-thermal plasma is its potential to oxidize particulates at low temperatures (Konstandopoulos 2013: 16). The typical reaction temperature is 200–300˚C, which is relatively lower than that of DPF regeneration by thermal oxidization (Kuwahara et al. 2012). A critical issue with this method is the requisite energy consumption and, therefore, relatively high fuel penalty (Kontandopoulos 2013: 16). Kuwahara et al. (2012) estimated that the plasma energy required for continuous DPF regeneration is approximately 5 % of the engine power output.

7.2 Wet Electrostatic Scrubber

Traditional particle abatement devices used in power plants and industry, such as water scrubbers, cyclones and electrostatic precipitators, are mainly designed and optimized to treat particles with sizes above or around 1 µm. Thus, they are relatively ineffective with the ultra-fine particles and nanoparticles emitted by diesel engines (Carotenuto et al. 2010). Moreover, they are also ineffective with gaseous CO, CO_2 and VOC emissions (Jaworek et al. 2015).

In order to decrease the emission of particulate matter in the submicron size range (<1 µm), wet electrostatic scrubbers (WES) have been developed. A wet electrostatic scrubber is a spray tower equipped with an electrified spray unit for the generation of electrically charged water droplets, and a particle pre-charging unit, as shown in Figure

15. A spray of charged water droplets (collectors) is dispersed in the scrubber chamber, and the electrostatic forces among positively charged droplets and negatively charged particles lead to rapid and effective particle capture. (Jaworek et al. 2013; Di Natale et al. 2015.)

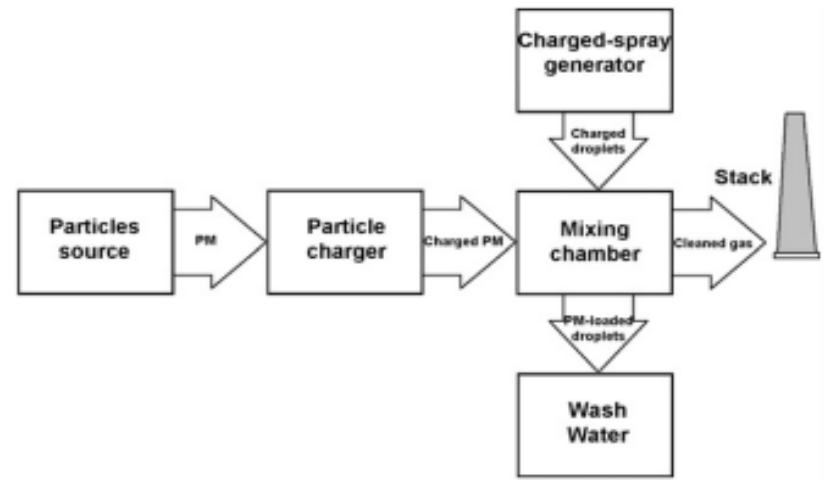

Figure 15. Schematic of wet electrostatic scrubbing system (Jaworek et al. 2013).

Wet electrostatic scrubbers inherit the structural and process advantages of conventional scrubbers, such as low-pressure drop and simple operation. An additional advantage of electrostatic scrubbers is their ability to also remove soluble gases. (Di Natale & Carotenuto 2015.) Experimental results published in the literature show that the removal of solid particles by water droplets is more efficient than with other mechanical methods. Experimental studies have also indicated that the energy consumption of the WES system is significantly lower than that of conventional scrubbers: an electrostatic scrubber requires 4–7 times less water than a conventional scrubber to remove the same amount of PM and SO_x. (Jaworek et al 2015.)

An innovative WES system for diesel particulate matter capture at pilot scale was recently developed and tested within the activity of the DEECON project (Innovative After-Treatment system for Marine Diesel Emission Control). Trial tests with a mono-cylinder diesel engine test rig indicated that the WES unit is capable of removing

particles as fine as 10 nm. The reduction efficiency of total particle concentration in the range 10–500 nm was 70-95 % in number (Di Natale & Carotenuto 2015).

Regardless of the simplicity of the physical background of wet electrostatic scrubbing, a number of technical problems have to be solved for the commercialisation of these devices. One of these problems is the charging of submicron particulates to sufficiently high levels to induce their motion towards the collectors, and the maximization of the charge on droplets. Another practical problem is the selection of material in order to avoid corrosion of metal elements, including electrodes, and problems with insulators cleaning. (Jaworek et al. 2013.) A fundamental drawback of all water-based techniques, including WES, is that wash-water treatment units are required to minimize liquid discharges and ensure that they are harmless to the environment (Di Natale & Carotenuto 2015).

7.3 Selective Catalyst Reduction and Filter

Meeting future emission regulations for inland navigation may require the use of both diesel particulate filter technology and the NO_x reduction catalyst SCR. Typically, the reduction of NO_x and PM emissions is performed by separate devices, as seen in Figure 16 (Tronconi et al. 2015). Such sequential system configurations, however, have several drawbacks:

1) Large volume of the whole exhaust cleaning system
2) Insufficient temperature for SCR catalyst during cold start when DPF is placed in front of the SCR
3) Unfavourable conditions for passive soot regeneration – lower temperature and lower NO_2 – if SCR is placed upstream of the DPF. (Johansen 2015.)

In order to solve these problems, catalyst manufacturers have recently showed a lot of interest in the development of combined SCR-DPF devices, often referred to as SCRF®, WSCR, or SDP, in which the porous walls of the DPF are saturated with SCR

catalytic material (Tronconi et al 2015). Experiments with SCR on filter integrations of vanadium- and Cu-zeolite based catalysts have shown promising results with high NO_x conversion capabilities and high (800˚C) thermal stability (Johansen 2015). A schematic drawing of SDPF layout is shown in Figure 17. The NO_2, which is generated in the DOC, is used both the continuous soot oxidation and the SCR reactions.

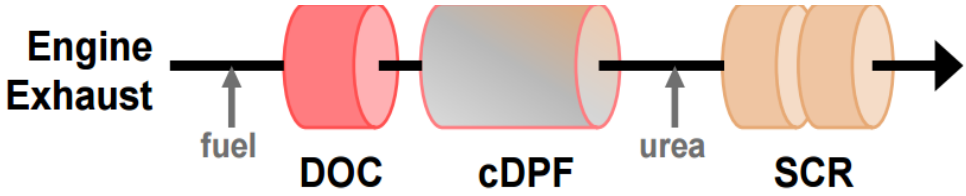

Figure 16. Typical exhaust system layout (Rappe et al. 2012).

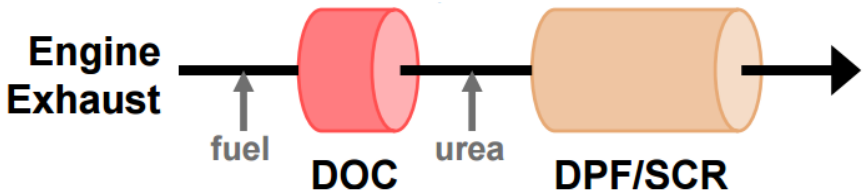

Figure 17. Possible future exhaust system layout (Rappe et al. 2012).

With SDPF systems, a number of benefits may be obtained:

1) Lower volume of exhaust system, lower weight, and possible lower cost
2) Improved heat transfers for both soot and NO_x conversion
3) Improved transport of gaseous components to catalytic surface
4) Earlier urea injection with improved cold start NO_x reduction performance. (Johansen 2015.)

The SDPF may also be used to replace the DPF in a DOC + DPF + SCR system to increase effective SCR volume without changing overall packaging, for example to enable an existing system to emerge with higher NO_X emissions resulting from

calibrating the engine for lower CO_2 emissions and higher fuel economy (Watling et al. 2012).

An important issue for the viability of the SDPF concept is the cross-interaction between the SCR and DPF functions. The fast SCR reaction $4NH_3 + 2NO + 2NO_2 \rightarrow 4N_2 + 6H_2O$ competes for NO_2 with passive soot regeneration reactions. (Johansen 2015.) As the NO_x reduction by SCR occurs much more rapidly than the NO_2–soot reaction, there is less NO_2 available to react with soot, leading to slower regeneration (Watling et al. 2012). In experiments performed by Tronconi et al. (2015), the influence of SCR reactions on passive soot oxidation were shown to be quantitatively prominent, and therefore this concept has to be carefully considered especially in applications which rely on continuous regeneration strategies.

Another aspect of the competitive interactions taking place in SDPF is the effect of soot presence on NO_x conversion. The reaction of NO_2 with soot to form NO alters the in situ NO_2/NO_x ratio, which is known to be a controlling factor in the NO_x reduction performance. (Tronconi et al. 2015.)

Taking into account these cross-interactions, the integration of the SCR and DPF functionalities into a single after-treatment device poses a major technological challenge which requires further extensive experimental work. An additional challenge of using this concept is the trade-off that needs to be made when choosing the optimal wash-coat loading to find a balance between NO_x reduction performance, filtration efficiency and the pressure drop behaviour (Tronconi et al. 2015).

The above-mentioned integrated systems for simultaneous control of several air pollutants seem to be extremely interesting options for exhaust gas after-treatment in the future, and the development status of these technologies should be followed. However, this study targets possible prototype testing in the end of 2016 or the beginning of 2017, and focuses on commercially ready technologies for particulate matter control. In the next chapters, an evaluation of different supplier technologies and solutions is made.

8 FILTER SYSTEM FOR 1 MW MARINE DUAL FUEL ENGINE

The purpose of this study was to find an effective and reliable exhaust gas after-treatment solution for particulate abatement for 1 MW marine dual fuel engine, both in gas and in diesel fuel use. The efficiency of the system has to be high enough to meet the PM emission limits specified in the proposed EU Stage V emission standard for inland waterway transport in Europe.

8.1 Technical Data of the Engine

The engine exemplified in this study is a 6-cylinder, four-stroke, medium speed dual fuel engine with direct injection of liquid fuel and indirect injection of gas fuel. As a dual fuel engine, it can be run in either gas mode or liquid-fuelled diesel mode. In gas mode the engine works according to the lean-burn Otto principle, with a lean premixed air-gas mixture in the combustion chamber. The gas is fed into the cylinder in the air inlet channel during the intake stroke. Instead of a spark plug for ignition – normally used in lean-burn gas engines – the air-gas mixture is ignited with a small quantity of pilot diesel fuel. The quantity of pilot injection is typically less than 1 % of full-load fuel consumption. This high energy source ensures reliable and powerful ignition of the lean air-gas mixture. In diesel operating mode, the engine operates only on liquid fuel oil. In diesel mode the engine can be run both on diesel oil or heavy fuel oil. In diesel operating mode, the engine works according to the normal diesel concept: liquid fuel is injected into the cylinder at high pressure and the fuel is ignited instantly due to the high temperature resulting from the compression. (Woodyard 2009: 42–44.) An example of the technical main data of a 1 MW marine dual fuel engine data is given in Table 6.

Table 6. Technical main data of the engine.

Cylinder bore	200 mm
Stroke	280 mm
Piston displacement	8.8 l/cyl
Cylinder configuration	6 in-line
Speed	1200 rpm
Engine output	1100 kW

In diesel operation mode the ultra-low sulphur diesel fuel EN590 (<10 ppm S, ash content <0.01 %) is used. The estimated lube oil consumption is 0.5 /kWh. The ash content of the lube oil is 1.5 %.

The maximum exhaust mass flow at full load in diesel mode was assumed to be 2.3 kg/s with exhaust temperature 340˚C. In gas operation mode, the maximum exhaust mass flow at full load 1.8 kg/s with exhaust temperature 360˚C was applied.

8.2 Requirements for the Particulate Filtering System

Based on the PM emission measurements of the engine, the target reduction efficiency for exhaust after-treatment system is >90 % for PM and >99 % for PN in diesel operation mode. Thus, the highest efficiency wall-flow particulate filter is required.

Other targets for particulate filtering system were defined as follows:

- Low pressure drop and parasitic load. Maximum pressure drop over the filter system 30–40 mbar.
- High availability, i.e. only little maintenance is needed. Ash cleaning interval >4000 h.
- Filter warranty 10,000 hours.

8.3 Conduct of the Inquiry

In order to find an optimal DPF-system for 1 MW dual fuel marine engine, numerous companies active within the exhaust gas after-treatment field were reviewed. The particulate filtering suppliers were found by surveying literature and from the Internet. A specific inquiry was sent to various potential suppliers for an evaluation of their technologies and solutions.

The suppliers were asked to specify the following information on their particulate filtering systems in their replies:

- Design and the size of the filter system
- PM and PN removal efficiency
- Pressure drop of the system
- Soot regeneration method
- Expected ash cleaning interval and estimated costs of the filter maintenance
- Price and delivery time
- Warranty terms

They were also asked to give information about the following topics:

- Is the system scalable for larger, e.g. 2 MW or even larger, engines?
- Would the design of the DPF for gas mode only be much different?

The response rate for the inquiry made for suppliers was 62 %. The following types of particulate filtering systems were proposed:

- passive systems with catalytic coated ceramic wall-flow filter elements
- passive system with DOC + uncatalysed ceramic wall-flow filter element
- passive systems with DOC + catalytic coated ceramic wall-flow filter elements
- passive-active system with catalytic coated ceramic wall-flow filter element + full-flow fuel burner

An overview of the proposed filter types and ranges of the key parameters are presented in Table 7.

Table 7. Overview of the proposed filter systems.

Filter types	wall-flow cordierite, wall-flow SiC, DOC + wall-flow cordierite, DOC + wall-flow SiC
Regeneration methods	catalytic coating, DOC + catalytic coating, catalytic coating + burner, NO_2 from DOC
Reduction efficiency	
• PM (%)	90-97.9
• PN (%)	97-99.5
Pressure drop	
• clean (mbar)	20-40
• operation (mbar)	30-73
Ash cleaning interval (h)	1000-5000

8.4 Comparison of the Proposed filtering Systems

The most important issues that have to be taken into account when choosing a filter system include filtration efficiency, reliability and durability of the filter system, regeneration method, maximum pressure drop over the filter, and maintenance requirements.

The filtration efficiency was in accordance with the targets (>90 % for PM, >99 % for PN) in most cases. One supplier reported slightly lower filtration efficiency for particulate number (97 %).

Both cordierite and silicon carbide filter elements were proposed. Cordierite is characterized by a very low thermal expansion coefficient and high resistance to

extreme thermal cycling. The disadvantage of cordierite compared to silicon carbide is a lower melting point, which is considered insufficient under certain uncontrolled regeneration conditions. In contrast, silicon carbide is characterized by a superior thermal durability. The demerits of silicon carbide are a higher thermal expansion coefficient (less resistant to extreme thermal cycling) and higher cost.

A number of fully passive systems were proposed. The advantage of passive systems is that no external energy is needed for the regeneration process. Regeneration at lower temperatures also reduces thermal stress, resulting in a longer life span of the filter media. A critical issue with fully passive systems is that passive filters require operating temperatures high enough to initiate combustion of collected soot. Lower loads and exhaust temperature may lead to poor regeneration and higher pressure drop. The minimum regeneration temperature for the proposed passive systems were 200–340˚C, and met the given minimum full load exhaust temperature 340˚C (in diesel mode).

A noble metal based catalyst – a DOC or a catalyst coating with precious metal – provides the additional advantage of a high conversion rate for CO and HCs. On the other hand, formation of toxic components or secondary particles which were not originally in the exhaust gas, so-called secondary emissions, has also to be kept in mind. It is known that catalytic converters containing platinum may increase the NO_2 emissions. (Weltens & Vogel 2008: 221, 225.)

A passive-active system was also proposed. The advantage of this method is that it is less dependent on exhaust gas heat. The regeneration can be triggered to occur actively with light loads and lower exhaust temperatures. The demerits of this kind of system are increased fuel consumption and increased thermal stress during active regeneration periods, higher maintenance requirements, and more complex and expensive control units.

A very important parameter is the maximum pressure drop through the exhaust system, caused by the hydraulic resistance of filtering system components (Majewski 2015c).

The pressure drop over the filter system should stay below the maximum allowable engine back pressure specified by the engine manufacturer. In this study, 30–40 mbar was applied. The maximum pressure drop across the filter system was ≤40 mbar in all alternatives at clean conditions. Some suppliers reported higher back pressure levels, up to 73 mbar, during engine operation, i.e. with some soot and some ash load.

The targets for DPF included high availability, i.e. only little maintenance should be needed. The ash cleaning interval was expected to be >4000 hours. The suppliers reported surprisingly large differences in expected ash cleaning intervals: 1000–5000 hours. Differences in ash storage capacities may be explained, to some extent, by cell designs. Filter designs with asymmetric cells in which the inlet cell is larger than the outlet cell have demonstrated a considerable increase in ash storage capacity for the same nominal filter volume, by simply increasing the size of the inlet cells to accommodate the ash (Sappok 2016). Also, some differences were found in calculating methods. Some suppliers informed that they can offer an ash cleaning service.

Generally, passive particulate filter systems do not require frequent maintenance, except for ash removal. Some suppliers stated that after approximately 10,000–12,000 hours the cartridges must be removed and maintained (renew coating). Warranty times varied between 8,000–12,000 hours or, respectively, 1–2 years.

The suppliers were also asked to specify if the DPF-system was significantly different in gas operation only. In general, the suppliers informed that the design of the filtering system would not be significantly different for gas mode only. The permissible maximum pressure drop over the filter system is the primary parameter here. Some suppliers proposed two differently sized filter systems, both for dual mode and gas mode. Most systems were said to be scalable to higher engine power, but bigger engines may require some special attention.

9 CONCLUSIONS

The objective of this study was to explore available after-treatment methods for the removal of particulate matter emissions from exhaust in inland navigation. A number of companies active within the exhaust gas after-treatment field were contacted and asked to send a proposal for the particulate filtering system for a 1 MW dual fuel engine. Several well-prepared proposals were received. The response rate (62 %) was sufficiently high to allow generalizing of the results. Based on the replies, the following conclusions and guidelines for the selection of particulate filtering systems for inland waterway vessels can be made.

Passive DPF systems with catalyst coating and/or upstream DOC can be regarded as the primary choice for particulate filtering systems in IWT applications. The advantages of fully passive systems are their low operational and low maintenance requirements. These passive systems also showed low operational costs, because no external energy is applied in the regeneration process. In general, passive systems are also relatively inexpensive. On the negative side, regeneration with fully passive systems is dependent on exhaust heat. However, with adequate catalyst formulation and/or with DOC upstream of the filter, the regeneration temperature can be adjusted to meet the minimum temperature of the exhaust.

When assessing the proposals, special attention was paid to high filtration efficiency (>90 % for PM, >99 % for PN), adequate regeneration temperature, high availability, and low back pressure. The pressure drop should stay below 40 mbar also during normal engine operation. Low pressure drop seems to be critical for many solutions, but some proposals meet the targeted 30–40 mbar also in saturated conditions. However, in most cases the pressure drop can be tailored by using the correct filter volume or by installing more filters.

In addition to fully passive systems, a passive-active filtering system was proposed. The advantage of catalysed filters with active regeneration support is that they are less dependent on exhaust gas heat. The regeneration can be triggered to occur actively with

light loads and lower exhaust temperatures. On the negative side, the demerits of CDPF + burner systems are linked to increased fuel consumption and increased thermal stress with active regeneration periods, higher maintenance requirements, and more complex and expensive control units. However, catalysed filters with active regeneration support in low temperature conditions can be seen as the most reliable method for particulate emission reduction. This alternative also had low pressure drop during engine operation, long ash cleaning intervals, and high reduction efficiency.

Noble metal based catalysts – a DOC and/or a catalyst coating with precious metal – provide the additional advantage of high conversion rate for gaseous carbon monoxide and hydrocarbon emissions. Platinum group metal (PGM) free catalysts, such as mixed metal oxide catalysts, showed lower activity for the oxidation of gaseous CO and HCs. PGM free catalysts also required higher regeneration temperatures compared to noble metal based catalysts. On the other hand, attention should also be paid to secondary emissions. It is known that catalytic converters containing noble metals may increase NO_2 emissions.

Finally, we can discuss the limitations concerning this study. It should be underlined that the above mentioned guidelines for the selection of particulate filter systems are limited to applications with ultra-low-sulphur fuel. As the sulphur content of diesel fuel is critical to DOC applications and noble metal catalysts, the applicability of these methods in, e.g., marine applications with high sulphur fuel is restricted. An interesting topic for further research could be the effect of high sulphur content in the fuel on filter design and filter regeneration.

For further research, a study of the effect of increased backpressure on engine performance and fuel consumption is also recommended to provide the necessary information for comprehensive operating cost analysis of particulate filtering systems. In addition to this, further research on combining diesel particulate filters with other after-treatment devices, primarily with SCRs, is recommended. The development status of integrated systems for exhaust gas after-treatment should also be followed.

10 SUMMARY

Diesel particulate matter is an extremely complex mixture of organic and elemental carbon, acids, such as sulphates and nitrates, metal ashes, particle-bound water and soluble organic compounds. Numerous epidemiologic studies in recent years have consistently linked particulate matter emissions in the ambient air to negative health effects for exposed populations. One of the main findings is that the size of particles is a crucial factor behind the observed health effects.

Nearly all diesel particulates have sizes of significantly less than 1 µm. As such, they represent a mixture of fine, ultrafine, and nanoparticles. Compared to larger particles, they can be breathed more deeply into the lungs, remain suspended for longer periods of time, penetrate more readily into indoor environments, and are transported over much longer distances.

Diesel particulates have been subject to diesel emission regulations worldwide. In September 2014, the European Commission proposed a new regulation on requirements relating to emission limits and type-approval for internal combustion engines for non-road mobile machinery, including inland navigation vessels. The proposal includes the introduction of a new emission stage (Stage V). This stage includes a number of important changes. It widens the scope of regulated engines, strengthens the emission limits, and furthermore, adopts particle number emission limits for several categories. The particulate number limits can be seen as a tool to limit ultrafine particulates from diesel engines, which are known to play the largest role in affecting human health.

These new emission limits will eventually require many ships to apply efficient exhaust gas after-treatment technology. The objective of this study was to explore available after-treatment methods for the removal of particulate matter emissions from exhaust. The main target was to find out what kinds of exhaust gas after-treatment solutions for marine dual fuel engines could meet future tightening PM emission standards in inland waterway transport in Europe, both in gas and diesel fuel use.

In the first part of this study, a literature review was conducted. This first part serves as a general description of different exhaust gas after-treatment technologies for particulate emission control, including catalytic converters, particulate filters, and auxiliary systems for supporting filter regeneration. Among these techniques, diesel particulate filters with high filtration efficiency and good regeneration characteristics were considered the best option. In addition to the above-mentioned conventional techniques, integrated systems for simultaneous control of several air pollutants were overviewed.

In the second part of the study, an evaluation of different supplier technologies and solutions were made. In order to find an optimal after-treatment system for a 1 MW marine dual fuel engine, several companies active within the exhaust gas after-treatment field were reviewed. The particulate filtering suppliers were found by surveying literature and the Internet. The targeted properties for particulate filtering systems were defined and a specific inquiry was sent to potential suppliers. The inquiry contained information about the targets for the particulate filtering system, engine data, and information about the fuels and the exhaust gas parameters. The suppliers were asked to specify the following items in their replies: design and the size of the system, PM and PN removal efficiency, pressure drop over the filtering system, soot regeneration method, maintenance requirements and the expected ash cleaning interval, warranty terms, price and delivery time.

Well-prepared proposals for particulate filtering systems suitable for 1 MW marine dual fuel engines were received from several suppliers. The received proposals were assessed and compared with the targets. A high emission reduction efficiency, high reliability, low pressure drop over the filter system, and high availability were considered the most important factors. Based on the proposals, the following conclusions were drawn:

1) Diesel particulate filters with catalyst coating and/or an upstream diesel oxidation catalyst can be regarded as the primary choice for particulate filtering system in IWT applications.

2) Catalysed filters with active regeneration support in low temperature conditions can also be seen as an advisable solution for particulate emission reduction.

Further research on the effect of high sulphur content in the fuel on filter design and filter regeneration is recommended. In addition, further research on combining diesel particulate filters with other after-treatment devices, mainly SCRs, is needed. Also, the development status of integrated systems for exhaust gas after-treatment should be followed.

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