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Critical Issues in Assessing Occupational Exposure to Diesel Dust Exhaust

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Summary

The Italian Interministerial Decree of February 11, 2021, introduces the diesel engine exhaust (DDE) among the carcinogenic occupational compounds, also establishing an occupational exposure limit. Elemental carbon (EC), improperly called black carbon, has been proposed as a tracer of DDE exposure; EC is the carbon that is quantified in the ambient matrixes after all the organic carbon has been removed; traditionally, EC is measured with a thermo-optical analytical technique. EC determination and relative interpretation are challenging for the following reasons: (i) the scarce availability of equipped laboratories hampers EC analysis, (ii) EC interpretation is not easy due to the lack of reference values. Finally, (iii) the limit value of 0.050 mg/m^3 of EC in the workplace appears too high compared to recently published exposure data. All these aspects stimulate a reflection on the significance of EC data in the context of both occupational hygiene and occupational medicine.

1. Introduction

Today, diesel engines are still widely used because they are efficient, durable, and with low maintenance, especially for heavy vehicles. They have been used on a large scale since the 1930s, first in mines and then in railway locomotives. For heavy vehicles, they appeared on the market in 1950 and became dominant between 1960 and 1970 [1]. Fire engines have been equipped with diesel engines since the 1960s, but the first measures taken to reduce exposure in car workshops date back to the 1980s, as reported in the IARC Monograph on "Occupational Exposure as a Firefighter" [2]. Since then, diesel engines have been widely used in various industries: transport, construction, agriculture, marine, manufacturing and mining, to power various vehicles, equipment and machinery. The most studied sectors for this type of exposure in scientific literature are mines and underground works [3]. Instead, the starting point for assessing occupational exposure to Diesel Dust Exhaust (DDE) is 2012, when IARC classified DDE as a Group 1 carcinogen [4]. Previously, it was considered a possible carcinogen. Data from the European Union's Roadmap on carcinogens [5] shows the scale of occupational DDE

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exposure: (i) more than 3.6 million workers in Europe are exposed to DDE; (ii) almost 4700 cases of lung cancer and over 4200 deaths are reported each year; (iii) workers who are frequently exposed to DDE in the course of their work have a 40% increased risk of developing lung cancer.

The assessment of exposure to DDE follows the International Labour Organization's (ILO) three-step process [6]: (i) hazard identification, (ii) workplace exposure assessment, (iii) identification of operating conditions and risk management measures (RMM) to control the risks.

Regarding the exposure assessment to DDE in Italy, a key milestone is the publication of the Interministerial Decree of February 11, 2021 [7], which implements EU Directive 130/2019 [8]. This decree adds "Oils previously used in internal combustion engines" and "exhaust emissions from diesel engines" to Legislative Decree 81/08, increasing the number of processes involving exposure to carcinogens from six to eight ("crystalline free silica" being the last included in 2020 by Legislative decree 44/2020) [9]. At the same time, the Interministerial Decree of February 11, 2021, introduces into the Legislative Decree 81/08 an occupational exposure level (OEL) for DDE, expressed as an airborne concentration of 0.05 mg/m^3 in elemental carbon (EC). This OEL value came into force on February 21, 2023, except for underground work, which will come into force in 2026. The choice of an OEL expressed as EC is not based on the toxicological properties of this element but rather on the fact that EC is the main diesel exhaust component [10]. The existence of an OEL for carcinogenic DDE means that this value should never be exceeded and, in this case, work activity must be stopped. Therefore, exceeding the limit value has important implications for the work activity, as well as for exposure control and health surveillance. The reference for discussing in detail the reasons behind the choice of the current occupational exposure limit must be sought in the official opinion of the SCOEL 2017 [11], reporting that it is not yet possible to establish "a critical threshold that could serve for the derivation of an OEL". Information provided by IARC also confirms that it is not possible to establish a critical health-based threshold for DEE but, in turn, this Agency suggests using EC, a significant percentage of DEE emissions, as an exposure indicator (as reported in art. 16 Directive 130/2019) [8].

Nevertheless, the choice of EC as a tracer involves several critical issues for its complex quantitative determination since EC is the carbon obtained by thermal volatilization under the flow of an inert gas, followed by oxidation to carbon dioxide (CO_2). Moreover, several critical issues are known for quantitatively assessing DDE exposure in different workplaces [12, 13]. Considering that diesel particulates contain carbon as EC and black carbon (BC), defining which particle size fraction best suits diesel exhaust sampling in workplaces is also a priority.

In light of the above considerations, this article analyzes these critical aspects, trying to explore the possible impact of the application of the Italian Interministerial Decree on February 11, 2021, on the practices of occupational hygiene and occupational medicine.

2. OCCUPATIONAL EXPOSURE TO DDE

Table 1 shows some work activities and/or macro-sectors with possible different types of exposures related to work environment and/or specific

Table 1. Some environmental and work occupational exposures to diesel dust exhaust.

Occupational sectors	Workshops	Industries	Constructions
Indoor	Mechanics	Compressor / Generator Operators	Miners
	Firefighters	Waste Collectors	Masons / Builders
	Administratives	Maritimes	Airport Workers
Outdoor	Firefighters	Carpenters	Masons / Builders
	Taxi Drivers	Waste Collectors	Forklift operators
	Bus Drivers	Maritimes	Airport Workers
	Truckers	Forklift operators	

tasks [14], highlighting that occupational exposure can vary for the same type of work indoors or outdoors. Short-term exposure to DDE can cause irritation of the eyes, nose, throat, and lungs. Prolonged exposure may increase the risk of developing chronic respiratory diseases and, in particular, lung cancer [14, 15]. DDE (as solid particulate matter and gaseous pollutants) can induce and develop cellular inflammation in the upper and lower airways. Moreover, DDEs are responsible for pro-inflammatory and pro-allergenic effects [16].

3. THE DUST DIESEL EXHAUST

3.1. Diesel Fuel

Diesel fuel is a derivative of petroleum that contains hydrocarbons from nine to twenty carbon atoms. Aromatic hydrocarbons account for about 30% of the fuel, sulfur content is less than ten parts per million (ppm), and the percentage of polycyclic aromatic hydrocarbons (PAH) is less than 8% [17]. Diesel fuel, mixed with air (nitrogen and oxygen) at high pressure in the combustion chamber, ignites spontaneously due to the high temperature. The diesel engine uses heat to convert the chemical energy in the fuel into mechanical energy. The combustion process is imperfect; other pollutants are also produced in addition to carbon dioxide (CO_2) and H_2O. Several factors, including the air/fuel ratio, the ignition timing, the combustion chamber turbulence, the air and fuel concentration, and the temperature reached, cause incomplete combustion. Incomplete combustion products are modified and exposed to high temperatures, and “soot” is produced in greater quantities than in petrol engine exhaust. There are hundreds of chemical compounds in DDE, of which forty are known to cause cancer.

3.2. Chemical Composition of DDE

N_2 (67%) and O_2 (9%) are in the combustion chamber, and CO_2 (12%) and water (11%) are the combustion products in gas phases together with pollutant emissions (1%). Pollutant emissions are composed of carbon monoxide (CO), hydrocarbons (HC), nitrogen oxides (NOx), sulfur dioxide (SO_2), and particulate matter (PM). Diesel engines are lean burn. Indeed, CO and HC concentrations are low compared to NOx and SO_2 (CO is the product of combustion when hydrocarbons burn with oxygen deficiency, and HC contains many compounds: alkanes, alkenes, and aromatic hydrocarbons). NOx is a mixture of nitrogen monoxide (NO) and nitrogen dioxide NO_2 with a predominance of NO_2. N_2 does not react with O_2 in the combustion chamber, and it is expelled from the engine as N_2; but when the high temperature in the cylinders is reached (>1600 °C), a reaction occurs. The initial combustion product is NO, which is then oxidized in the atmosphere to NO_2. NO_2 is five times more toxic to the respiratory system than NO. Cars are the main source of NOx, with diesel engines accounting for 85%. After NOx, PM is in greater quantities compared to other pollutants, and it is made up of 31% to 41% of carbon [18] (Interministerial decree February 11, 2021, asserts that carbon must be quantified as EC). Engine diesel PM is six or ten times higher compared to gasoline. They are spherical particles with a diameter of 15-40 nm, and more than 90% of PM has a diameter of 1 micrometer. There are emission control systems for diesel engines (for cars, not aircraft). The Diesel Oxidation Catalyst (DOC) controls CO and HC emissions. At the same time, the Diesel Particulate Filter (DPF) is used to control PM emissions, and finally, the Selective Catalytic Reduction (SCR) control system is used to control NOx. Currently, there is no system for controlling SO_2, which is needed to reduce the sulfur content of diesel fuel. Despite these control systems, the emission of pollutants into the atmosphere has the percentages shown above. As shown in Figure 1, PM comprises a "solid carbon core" to which hydrocarbons are adsorbed, and liquid hydrocarbons appear near the adsorbed hydrocarbons. Adsorbed and liquid hydrocarbons are sometimes called the soluble organic fraction (SOF). Hydrated sulfate species, forming inorganic fraction (IF), are associated with liquid hydrocarbons. The solid carbon is sometimes called soot, but historically soot has been called BC [19].

Carcinogens such as benzene, formaldehyde, and PAH in DDE have been known for years [20]. We underscore the complexity of PAH mixtures being a complex mixture of compounds with different

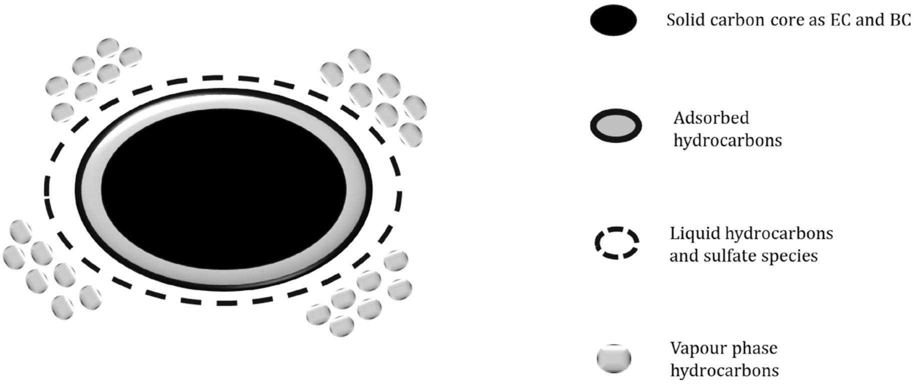

Figure 1. Schematic figure of particulate matter (PM) (amended by Martin V. Twigg and Paul R. Phillips, Platinum Metals Rev., 2009, 53, (1), 27-34).[19]

carcinogenicity, different concentrations, and possible formation of secondary toxic compounds. In addition, there are several non-carbon-based compounds, such as arsenic, cobalt, chromium, mercury, nickel, and phosphorus compounds, and organic compounds, such as acrolein, acetaldehyde, xylenes, etc. Because of the large number of chemical compounds present in DDE (including carcinogens), it would be useful to find more specific markers that do not have a background as EC (marker whose quantification is required by Interministerial Decree 2021). In addition, as reported in the Copenhagen Airport case study [21], caution must be exercised concerning the size of the engine particles. The study reports a dramatic scenario: "The result will be inhalation of 500 million particles per minute. This equates to 240 billion ultrafine particles per workday, a significant proportion of which are deposited in the most critical parts of the lungs (the alveoli)" [21]. This data shows that the number and the size of particles are important, and probably, they cannot be replaced by the measurement of EC alone. Moreover, the chemical composition of DDE has changed over time, from traditional diesel engines to those with new technology. Precisely in the new ones, EC decreases (from 75% to 13%) and organic carbon (OC) increases (from 19% to 30'%) [11]; also, NOx, CO, HC, and PM decrease in new engines diesel [22]. In particular, Piia Taxell reported that "for the new technology, diesel engine exhaust with significantly reduced particle mass and EC concentration, EC may not be an equally useful marker" [23].

3.3. Size Distribution and Mass Dust Content of Diesel Engine Exhaust. What Implications for Risk Assessment?

Exhaust emissions from diesel engines are a complex mixture of gas, vapors, and aerosols (all states of matter are present). PM or Diesel Particulate Matter (DPM) or Diesel Exhaust Particulate (DEP) are solid particles emitted by diesel engines as part of the DDE. In the IARC Press Release 213 of 12 June 2012 [24], some studies considered complete diesel emissions, which caused an increase in the incidence of lung cancer in rats. These studies showed that (i) the gas phase (with particles removed) did not increase the incidence of respiratory cancers in any of the species tested; (ii) the particulate phase caused malignant lung tumors in rats and sarcomas at the site of injection in mice.

The size distribution of particles emitted by diesel engines varies according to engine type, engine operating conditions, fuel formulations, lubricating oil, additives, and emission control systems. Substantial differences are also found depending on the age of the engines: the newest, compliant with Euro IV-VI, have a different emission composition from that of conventional diesel engines [11].

Diesel engine exhaust emissions are mixtures of hundreds of chemical compounds, which are emitted partly in the gaseous phase and partly in the particulate phase [25]. Particle mass is reduced by more than 90% in the case of Euro IV-VI engines compared to Euro I and II engines [23]. Concerning the size distribution of the emitted particles, a recent study reports that the particles containing OC and EC peaks at 330–550 nm, with OC/EC ratio showing two peaks in the ultrafine (< 100 nm) and accumulation modes (170–330 nm) [26]. Moreover, in the study of Eric Garshick et al. [10], EC in PM1 was tested using the NIOSH 5040 method [27], concluding that diesel emissions contributed significantly to the EC in PM1 in the United States in urban areas before 2006. Fine and ultrafine organic particles, which can penetrate the respiratory system, are also reported as produced by diesel engines and aircraft engines following incomplete combustion [28] [21]. Aircraft engines emit 1000 times more particles per kg of fuel than modern diesel engines (EURO V/VI); this is an important critical point in these workplaces, and an example of such a case of occupational exposure can become a public health problem.

Therefore, without prejudice to SCOEL's opinion on differences in diesel exhaust, the IARC emphasizes the need not to make a distinction between age and diesel engine type for the exposure risk since the qualitative and quantitative composition of emissions depends on the type and age of the engine, the emission control system, the development, maintenance, and mode of use.

As a final consideration regarding the age of diesel engines, the Italian Interministerial Decree of February 11, 2021, adds "diesel engine exhaust emissions" as carcinogenic compounds without considering any differences in their characteristics or age. On the other hand, the particle size fraction to be selected in diesel exhaust sampling for EC determination remains an open question. If this is not specified as notation to the limit value, industrial hygiene practice requires sampling the inhalable fraction, even from a precautionary perspective. The NIOSH 5040 method would fully comply with this practice, requiring sampling of the inhalable fraction with a three-piece cassette [27].

4. Exposure Assessment by Comparison with OEL

4.1. Sampling of Elemental Carbon

The carbon components of PM (EC, OC and BC) take different names based on the availability of different measurement techniques. Also, different terms refer to the same type of exposure to carbon in PM, particularly in the fine fraction. In this regard, the Italian legislation does not indicate which dimensional fraction of the particles have to be sampled in assessing the exposure, but a technical document published by the Italian Association of Industrial Hygienists (AIDII) in March 2023 provides guidance, as far as the technical standards are concerned. The AIDII guide [12] refers to:

a. STANDARD UNI EN 14530/2005 [29] (Atmospheres in the workplace. Determination of diesel particulate matter. General Requirements). This standard defines EC as residual soot nuclei after removing OC from particulate matter. It provides for sampling the respirable fraction (according to the UNI EN 481:1994 standard) [30].
b. STANDARD UNI EN 16909/2017 [31] (Ambient Air – Determination of EC and OC deposited on filters). This refers to the UNI EN 12341:2014 [32] standard and indicates PM2.5 as the fraction to be sampled.

The UNI EN TECHNICAL STANDARDS suggest sampling respirable or fine fraction of PM, but certainly not the coarse one. The Italian Interministerial Decree of February 11, 2021, indicates EC as a marker for assessing DDE exposure. The most suitable method for occupational exposure to diesel exhaust seems to be the NIOSH 5040 (NIOSH Analytical Methods Manual, V Edition year 2016) [27]: in this method, soot is considered synonymous with EC.

The method involves personal samplers with 37 or 25-mm diameter quartz fiber filters. Filters should be heated for 1-2 h at ~800°C to ensure any removing contaminants. The sampling flow is between 2 and 4 L/min with a minimum volume of 142 L. Lower flow rates are used in dusty environments to avoid filter overloading (cyclone or impactor is recommended to prevent interference). The sampling respiratory zone for the workers corresponds to a hemisphere (radius 30 cm) extending in front of the human face; technical normative EN 1540:2011, ISO 18158:2016 [33] are used to define this area, but it is not selective of any particle size fraction. Some authors show that the EC is a better marker than gravimetric methods (not suitable for low concentrations (< 200 μg/m^3) of PM in air) [34], being it a more selective indicator of diesel engine emissions and representing a considerable fraction of the mass of PM.

In addition, it is stated that EC is a specific marker of occupational exposure to diesel engine emissions regardless of the particle fractions (inhalable and respirable fraction). The sampling of particle size fractions according to the UNI EN 481:1994 [30] standard is not considered necessary except in mining activities (where the sampling of the respirable fraction is recommended).

Nevertheless, some papers have been reviewed [35], providing airborne concentrations of EC in the workplaces in both inhalable and respirable fractions. Diesel engines emit many fine and ultrafine particles, as organic particles produce incomplete combustion [28]. Ultrafine particles (UFPs) are expressed in particle number concentration (#/cm^3) rather than mass concentration (mg/m^3). UFPs (< 0.1 μm in aerodynamic diameter), together with the nanoparticles (< 0.03 μm), contribute the majority of the particle number. It is also reported that the NIOSH 5040 method does not interfere with cigarette smoke or other carbon-based aerosols because these are mainly composed of OC. Still, once again, attention must be paid to the new diesel technology, where the percentage of OC increases and EC decreases [11]. These aspects and critical issues are not mentioned in the February 11, 2021, Interministerial Decree. Finally, to obtain sufficiently low limits of quantification (LOD) (less than 2 mg/m^3), it should be noted that more than 1 m^3 of air needs to be sampled. Therefore, long sampling times are usually required.

4.2. Analysis of Elemental Carbon

Several analytical thermo-optical methods (such as NIOSH 5040, NIOSH-like, EUSAAR2, IMPROVE) exist for the EC quantification. Briefly, the thermo-optical measurement involves two heating ramps: the first one, in helium gas, is used to determine the OC, whilst the second, at a higher temperature, in an oxygen oxidative atmosphere, is necessary to measure the remaining oxidized carbon. CO_2 released after the two heating ramps is quantified directly by the IR detector or after methane reduction with an FID detector. During thermal analysis, the instrument measures the laser transmittance at 660 nm through the filter to determine the split point between OC and EC. The difference quantifies elemental carbon: total carbon (TC) minus OC (EC = TC – OC).

Thermo-optical methods EUSAAR2, IMPROVE, NIOSH 5040, and NIOSH-like are four different thermal protocols for heating the sample with differences in the maximum temperature value of the first phase: IMPROVE (used in the USA) and EUSAAR-2 (used in Europe) are medium-low temperature protocols, in which the first phase ends at 550 °C (IMPROVE) or 650 °C (EUSAAR-2). In comparison, the two NIOSH protocols end the first phase at 870 °C (QUARTZ or NIOSH-like) or 850 °C (NIOSH 5040). In general, low- to medium-temperature protocols result in higher EC concentrations than those from high-temperature protocols. This may be due to an incomplete evolution of OC in the first phase of medium-low temperature protocols (underestimation of OC, leading to an overestimation of EC), or to a pre-combustion effect of EC in high-temperature protocols (underestimation of EC and consequent overestimation of OC) [36].

5. MAIN CRITICAL ISSUES

5.1. Elemental Carbon *vs* Black Carbon (EC *vs* BC)

The methods for EC quantification, based on comparative differences between OC and EC determinations in the same line measurement (IMPROVE,

Table 2. The pro and cons of EC and BC measurements.

	Elemental Carbon (EC)	**Black Carbon (BC)**
PRO	Good specificity High sensitivity	Immediate and continuous measurement
CONS	A long and laborious measure High measurement uncertainty	Possible interferents

EUSAAR2, NIOSH 5040, NIOSH-like), imply measurement uncertainty to be added to quantitation uncertainty. The quantification of EC is difficult because there are only a few laboratories that can perform this determination in industrial hygiene. Still, many are equipping themselves as environmental agencies did in the past. In addition, the term BC is often used as a synonym for EC, as reported in the NIOSH METHOD 5040 [27]. BC is unbound carbon as EC, but these commonly used terms are potentially ambiguous.

In 2011, the Global Atmospheric Watch-Scientific Advisory Group (GAW/WMO) suggested definitions, which can be summarized as:

a. EC is carbon measured with a thermo-optical analytical technique.
b. BC is carbon measured with an optical analytical technique.

BC and EC refer to materials with different optical and physical properties (quantification of carbon with two different analytical techniques) rather than compounds with well-defined properties [37]. The determination of BC consists of an optical absorption measurement at one or more predetermined wavelengths and not an actual measurement of chemically well-defined parameters. It is also evidenced that EC is used as a surrogate to assess exposure to diesel PM. The word "surrogate" is used in a paper by Birch and Cary [38]. Could we assume BC is less "surrogate" than EC for quantifying diesel exposure? However, neither the EU nor the Italian legislation mentions these aspects.

The choice of using EC as a tracer of DDE, which has been endorsed by all official methods and technical standards worldwide, is probably due to three factors: i) The carcinogenic mechanism of DDE is mainly due to the particulate fraction (rather than exhausted gases); ii) EC represents more than 80% of DDE released particles; iii) EC measurement is less biased by interfering factors (e.g. cigarette smoke/OC) in comparison with BC [27].

In addition, BC is a component of the PM, the reduction of which can lead to climate and pollution benefits. Knowledge of the quantities and emission sources involved is necessary for effective intervention. The International and national scientific community should focus on standardization and, in particular, on definitions regarding the carbonaceous fraction of particulate matter and of measurement standards. BC is a primary pollutant from diesel engine exhaust and its measurement is important, but this does not mean that measuring black carbon could be prematurely endorsed as a good indicator of occupational exposure to diesel engine emissions. Further studies are needed to compare EC and OC measurements in ambient matrixes from different workplaces.

Finally, on April 2024, the European Parliament adopted the text for the legislative resolution on the proposal for a directive of the European Parliament and of the Council on ambient air quality and cleaner air for Europe [39], in which new rules are set for several pollutants including particulate matter (PM2,5, PM10), NO_2 and SO_2. The air quality standards shall be reviewed by 31 December 2030, and they are also based on the revised World Health Organization (WHO) Air Quality Guidelines, in which black carbon/elemental carbon was also a concern [40].

The new EU Directive updates the last version of the Directive on air quality in Europe [41]. It suggests continuous monitoring of EC, BC, and OC in the air. It proposes air quality standards for different pollutants, but no standard for EC is set.

Therefore, a new question arises regarding occupational exposure to DDE: is measuring both EC and BC in workplaces better? BC measurement can indeed be considered complementary to determinations via the thermo-optical reference technique.

5.1.1 Health Effects of Black Carbon

BC influences climate and pollution and has adverse health effects [42]. In 2012, the World Health Organization (WHO) highlighted that BC and EC were strongly correlated and measured with different analytical techniques. Moreover, the WHO has evidenced that BC is a carrier of other toxic or carcinogenic compounds. After 2012, new articles [43-45] reported that BC was not only a carrier, but it had effects on cardiovascular events and premature deaths in humans. These BC health effects further confirm the need to quantify it, and the WHO, in the 2021 Air Quality Guidelines update, included statements of good practice to address concerns about the health and environmental effects of the BC/EC.

5.1.2 Analytical Quantification of Black Carbon

The quantitative measurement of BC is done with a multi-spectrum instrument that constantly measures the transmittance of light at ten different wavelengths (from "near UV" to "near IR"). The instrument calculates, in real-time, the concentration of BC through filter support on which particulate matter accumulates. The room sampling system has a heater, and a sample flow rate of 2 or 5 l/min can be set. The analyzer uses a very common and inexpensive filter and allows the choice of the filter belt feed mode according to the concentration of BC. The instrument has sampling heads that allow the alternative measurement of PM10, PM2.5, or PM1. Using aethalometers for BC monitoring, high temporal resolution exposure data can be collected, which is very useful for identifying potential high-exposure peaks linked to specific work activities and thus to set up an effective risk management strategy.

The BC measurement may have interfering substances. It is well known that, in situations where particles opaque to light radiation, such as crustal particulate matter and heavy metals, are present, significant interferences can potentially lead to a significant overestimation of actual exposure levels [46].

5.2. Workers Exposed and Unexposed

In Occupational Medicine, “Exposed Workers” means workers who have an exposure above the background levels of the general population. However, although the EC can be considered a ubiquitous contaminant, there are no standard or guideline values for the protection of the health of the general population and this gap does not help the assessment of occupational exposures to DDE. The recently published Guideline of the Emilia-Romagna Region [47] refers to updating the guideline to discriminate between “Exposed and Unexposed Workers” in the future.

5.3. Elemental Carbon Values in Different Workplaces

Several papers have been published using EC as a marker to quantify occupational exposure to diesel exhaust. Among them, the most recent paper of Plato [48] reports measurements carried out in 72 different workplaces that are assimilable to life environments such as buses and cars and for operators on non-road equipment over a long observation period, ranging from 1950 to 2005. In this study, only a few recent data come for direct measurements of EC, while for the most part, the exposure has been estimated by a model adjusted with indirect measures. Exposures for workers exposed in the above environments decreased over the years, resulting below the OEL of 50 $\mu g/m^3$ set by the 2021 Interministerial decree. However, it should be noted that this study was carried out in Sweden, where air pollution levels are different from those in Italy, thus making exposures in working environments similar to living environments not entirely comparable.

In Italy, we have some data of EC [49] and BC [50] for general population exposure, with values varying over the seasons [51]. As a general consideration, environmental exposure to the EC ranges between 0.01 $\mu g/m^3$ and 5.1 $\mu g/m^3$ for EC [52], a maximum value that is approximately 10 times lower than the corresponding OEL. This “background” value, together with the measured EC concentrations in different workplaces, published in the

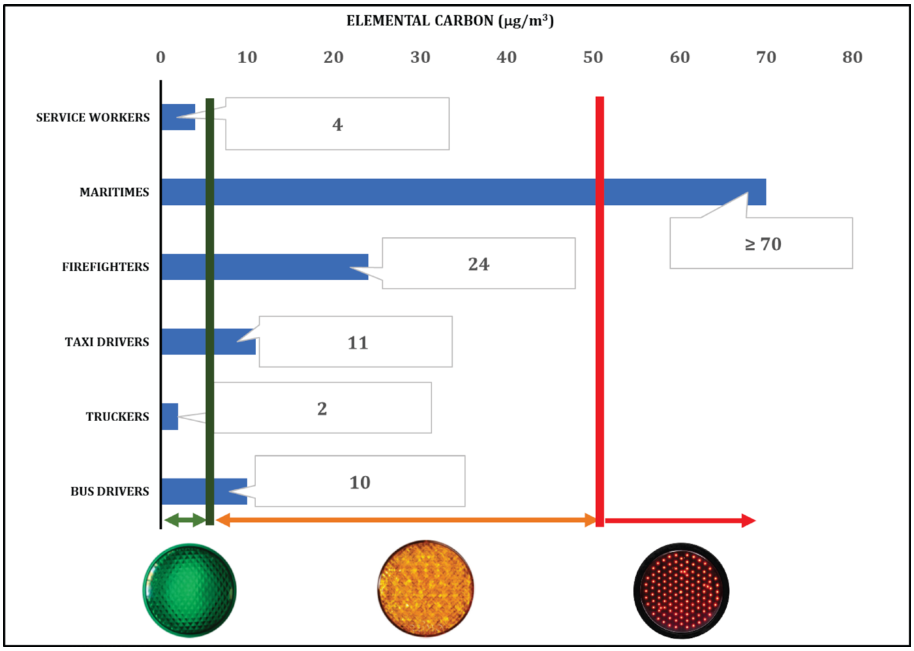

Figure 2. Mean air concentration for EC for exposed and unexposed workers (blue bars) **(Data from Pronk, 2009)** [35] and the general population (green line). The red line refers to the Italian OEL.

scientific literature for European and non-European workers [35], has been graphically compared with the OEL for EC in the graph shown in Figure 2.

Figure 2 shows that installers and truck drivers, so-called "unexposed workers," are exposed to EC values equal to or below the level for the general population; the exposed workers, instead, present exposure levels both below the OEL of 50 µg/m^3, taxi drivers, firefighters, bus drivers, and above 50 µg/m^3, maritimes. For these latter, a re-entry activity below the OEL is mandatory. This type of scheme may be useful in discriminating between exposed and not-exposed workers for the purpose of health surveillance for exposure to DDE.

5.4. Occupational Exposure Limit (OEL) for Elemental Carbon is not Health-Based

Another critical question is: "How precautionary are we in protecting workers from exposure to diesel engine exhaust fumes?" [53]. The 50 µg/m^3 limit value set by the 2021 Interministerial Decree is a regulatory, not health-based limit. It is a compromise between health and technically achievable values. The report by R. Vermeulen [53] shows the excess risk of lung cancer in Europe on the population of workers (229 million), setting different OEL values for exposure to DDE.

For an OEL value of 50 µg/m^3, an excess risk of 268 workers out of 10000 is obtained. For an OEL of 10 µg/m^3, the excess risk results in 166 workers out of 10000, which drops to 26 in the case of 1 µg/m^3. In the Netherlands, the Health Council, based on the exposure-response relationship by Vermeulen et al. [54], has set a health-based limit of 1 µg/m^3 for occupational exposure to DDE. To increase workers' health, the value of OEL should be reduced in the coming years, consistent with the technical possibility of measuring increasingly low concentrations.

6. CONCLUSION

This work highlights the critical issues in European legislation and, consequently, in the Italian one for assessing exposure to DDE. The 2021 Interministerial decree was issued by setting an OEL for EC about ten years after the publication of the 2014 IARC Monograph, where DDE was classified carcinogenic for the first time. From a scientific point of view, the new legislation is already non-exhaustive because BC (and eventually OC) can be quantified as a carbon-based marker for this type of exposure in addition to EC. In Italy, the Environmental Agencies have quantified EC, BC, and OC for years, as also required by the proposal to amend the European Union Directive of October 2022, which invited all Member States to carry out a greater number of quantitative determinations of the three markers to control air quality in Europe. The amended Directive does not set a standard value for EC, referring to the general population's exposure, while from February 2023, the occupational exposure limit of 50 μg/m^3 as EC is operative for workers. The 2021 Interministerial Decree also does not indicate the particle size fraction to sample in the workplaces for quantifying EC, BC, and OC. Still, it is known that particle size is an important parameter to consider for the risk of exposure because the smaller the size, the greater the probability that particles deeply penetrate the respiratory system, up to the alveoli. The Environmental Agencies show that all BC in the atmosphere is present in the sub-micrometric fraction of particulate matter (PM1).

To date, there is a lack of data in Italy on the environmental monitoring of BC and EC both in the workplaces and in the living environments. Moreover, the 50 μg/m^3 limit for EC seems already high in a more concrete perspective of safeguarding workers' health. Finally, the thermo-optical quantification of EC is a laborious multi-step method (and probably not useful for new technology diesel engine exhaust) with the risk of a high measurement uncertainty that can influence the exposure results with consequent impact also the health surveillance.

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