This report has been reviewed by the staff of the California Air Resources Board (ARB) and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the ARB, nor does mention of trade names of commercial products constitute endorsement or recommendation for use.
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1. Introduction

Due to the major contribution of motor vehicles to both air pollution and greenhouse gas (GHG) emissions in California, the ARB staff is proposing to establish new emission standards for light-duty vehicles sold in the State. Staff is in the process of developing regulatory standards for these emissions as part of the Low Emission Vehicle (LEV) III program. In so doing, California is simultaneously addressing local air pollution that is caused by criteria emissions and GHG emissions that contribute to climate change. There are two components of this new program, the LEV III-SULEV element, in reference to the Super Ultra Low-Emission Vehicle (SULEV) emission compliance designation, and the LEV III-GHG component.

ARB staff recognizes the nexus between air pollution and climate change and is, therefore, simultaneously confronting these two major environmental issues to balance trade-offs and to take advantage of synergies where possible. The proposed standards will promote further improvement in motor vehicle technology and continue California’s tradition of science-based, technology-forcing policy in the interest of air quality and now also the global climate. Advances in engine design, combustion technology, aftertreatment devices, and cleaner fuels, will reduce emissions and enable compliance with new emission limits. Engine and drivetrain developers will be expected to more efficiently harness the energy in the fuel and in the process emit less carbon per vehicle mile driven. Aftertreatment design will require innovation to ensure that criteria pollutant emissions are reduced to increasingly lower levels in conjunction with the new advanced combustion concepts that will yield improved efficiency. Because of this multiple-pollutant challenge, the new criteria and GHG emission standards are being developed concurrently to ensure that the automotive technologies, standard stringency, and timing of the standards are aligned. Particulate matter (PM) emissions are at the center of this multiple-pollutant challenge because of its link to air quality and climate change.

Staff has reviewed extensive research in various areas including measurement-based characterization of emissions, emission inventory, and health impact assessment, all related to light-duty vehicle (LDV) PM emissions. As a result, several distinct issues have emerged related to PM emissions and staff has determined that regulatory consideration may be warranted as discussed originally in the first “Preliminary Discussion Paper – Amendments to California’s Low-Emission Vehicle Regulations for Criteria Pollutants – LEV III.” This second discussion paper presents for discussion three distinct, but interrelated topics in the context of developments toward the LEV III standards. The topics are,

1. **PM mass emissions and the stringency of a proposed limit,**

2. **The number of particles in the emissions and an optional compliance limit based on solid particle number (SPN), and**

3. **The fraction of PM emissions that is black carbon (BC), a potent climate**
This report is structured following the three topics. After this introductory section, Section 2 provides a discussion of staff’s current testing of PM mass emissions and how results are informing the regulatory LEV III-SULEV program. The section provides information on PM mass emissions from different current and emerging gasoline engine technologies, potential options for reductions, amendments to the existing LEV II mass standards, and testing methods. The section provides an overview of the extensive work to date on internal combustion-generated ultrafine particle number emissions and the rationale behind the proposed option for the inclusion of a new SPN limit in the LEV III-SULEV program. Finally, Section 3 discusses the inclusion of BC in the LEV III-GHG standards for carbon dioxide and other GHG emissions. Control of BC has emerged as a new topic of high interest since it provides a means to mitigate some climate change effects in the short term.

The objective of this second discussion paper is to provide an update on the current status of staff’s work toward developing the technical basis for the proposed PM amendments and new standards in advance of the next public workshop. Staff is using this second preliminary discussion paper to again solicit additional feedback from stakeholders on all of the PM-related issues over the upcoming months of the ARB LEV III rulemaking process. In addition to the May 18, 2010 technical workshop that will include discussion of PM, subsequent ARB public workshops will also provide opportunity for stakeholder input, and stakeholders are encouraged to provide feedback to ARB staff through written and electronic submissions. Staff notes that this document and statements herein are preliminary. In addition to participating in the dialogue at the public workshop, industry groups and environmental stakeholders are invited to discuss the proposed regulatory provisions in face-to-face meetings with staff if necessary to protect confidential information.

2. Particulate Matter Emissions

The existing California LEV II regulations limit PM mass emissions from new LDVs to 0.010 g/mile at full useful life. Vehicle categories covered under the program include all passenger cars, light trucks, and medium-duty passenger vehicles. The current set of standards includes the emission category designations of LEV, Ultra Low-Emission Vehicle (ULEV), and SULEV. LDVs are responsible for only about two percent and three percent of PM10 and PM2.5 emissions, respectively. However, the relative fraction of PM contributed by LDVs could rise significantly because other sources of PM are becoming cleaner in response to requirements for new controls, and because LDV PM emissions could rise when new advanced gasoline combustion technologies for greater thermodynamic engine efficiency are more widely introduced.

The primary impact of the 0.010 g/mi standard has been to force the use of highly efficient diesel particle filters (DPF) on diesel-fueled vehicles, which typically have higher levels of engine-out PM mass emissions than gasoline-fueled vehicles. In practice, new gasoline-fueled LEV II vehicles, which predominantly use port fuel
injection (PFI) technology emit about 0.001 g/mile, or ten percent of the standard. This fortunate situation of overcompliance has given rise to a desire to sustain the same PM benefits from all future California LDVs as those afforded by the current clean PFI technology.

The U.S. Environmental Protection Agency (U.S. EPA) and the National Highway Transportation and Safety Administration (NHTSA) have recently adopted new national LDV GHG emission standards and corporate average fuel economy standards similar to ARB’s Pavley regulation adopted in 2004. The new national standards as well as the LEV III-GHG standards that are in development for potential adoption this year are expected to encourage wider adoption of technologies that reduce CO₂ emissions, such as Gasoline Direct Injection (GDI). However, GDI technology tends to have higher PM mass and particle number emissions than conventional PFI technology. Reports in the published literature point to GDI PM mass emissions in the range of 2 to 20 mg/mi

In a recent study conducted by ARB, staff confirmed that PM mass emission levels from some GDI vehicles approach the current LEV II standard. If not abated, the GDI combustion system has the potential to emit two to eight times more PM mass than PFI vehicles. To limit this backward slide in PM emission performance and the proportional contribution to the PM (and BC) inventories, the proposed LEV III-SULEV amendments incorporate a reduced emission limit for PM mass.

ARB staff is also proposing a new SPN emission limit as an alternative compliance option. The reason for inclusion of this new option is three-fold. The proposed SPN optional limit (1) provides flexibility to car makers subject to this rule who may wish to use, to the extent that is appropriate, the compliance information necessary for Europe, where a SPN standard is in place; (2) updates California’s policy for motor vehicles by formally recognizing the growing concern about exposure to small particles in urban air, and (3) recognizes particle counting as a measurement approach that is superior to the conventional gravimetric method for PM mass determination in terms of speed, precision, and ease of use. A SPN emission limit would prevent a regression in solid particle emission rates because as discussed later, GDI PM mass emissions consist mainly of solid carbonaceous particles or soot. The SPN standard provides an alternative means to demonstrate effective PM mass emission control using a measurement procedure that is simpler, faster, and less costly than the complex gravimetric methods for PM mass determination. As being considered by ARB staff, both proposed PM mass and SPN standards achieve a similar result by ensuring well-controlled combustion technology.

2.1 Health effects

The health impacts of ambient PM are estimated from epidemiological studies that show associations between ambient concentrations of PM mass and various health endpoint measures. For example, long-term and short-term epidemiological studies have found associations between PM2.5 and PM10 and increases in all cause mortality, stroke related deaths, and respiratory disease related deaths\(^4,5,6\). Although epidemiological studies with both PM10 and PM2.5 suggest that cardiovascular effects are associated with smaller particles, there are few reports that show that ultrafine particles (UFP) exposures lead to increased mortality. This lack of studies is not necessarily a reflection of UFP’s lack of toxicity but rather the dearth of monitoring data needed to accurately assess ambient air concentrations, and estimate exposures.

Despite this gap in epidemiological knowledge, there have been several studies that compare the physical characteristics, chemical properties, and biological activity among PM10, PM2.5, and UFP in both animal and in vitro cellular systems. Of particular interest is the difference in the ability of particles to induce aortic and coronary atherosclerosis in mice that are susceptible to atherosclerosis. In one of the few studies\(^7\), which compared both mass and particle number of PM2.5 and UFP exposures, mice were exposed to concentrated levels of either PM2.5 or UFP. The results showed that the UFP–exposed mice were more prone to aortic atherosclerosis when compared to mice exposed to PM2.5 or filtered air. This experiment illustrates that UFP are potentially more toxic than PM2.5 at least on a per mass basis and that particle number may be an important metric to consider for toxicity studies.

The possible increased toxicity of UFP may be explained by their relatively higher surface area when compared to both PM2.5 and PM10 particles. This greater surface area results in higher relative content (by percentage of total mass) of both organic and elemental carbon, and importantly, polycyclic aromatic hydrocarbons (PAHs) which have been shown to be cardiotoxic in treated animals\(^8,9\). In a comparative study of PM10, PM2.5, and UFP by Li and coworkers\(^10\), UFP not only had the highest content of PAHs by mass of the three types of particles but also displayed the highest biological activity associated with PAHs in their model cell systems. This included higher levels of oxidative stress biomarkers, as well as evidence of increased mitochondrial damage.

Currently, ARB is funding a project to determine how the toxicity of PM2.5 and UFP varies depending on the level of the semi-volatile fraction in the particles emitted from vehicles and other sources.

The toxicity of UFP may also be linked to the fact that they deposit in the alveolar regions of the lung and can be transported into the circulatory system. This process is thought to ultimately increase systematic concentrations of UFP and/or their associated chemicals, which may affect other organs or lead to increased systematic inflammation.

There have been a limited number of studies of the health effects of UFP near roadways. For example, a recent study by Cho et al\textsuperscript{11} compared the toxicity of size-fractionated PM collected at different distances from an urban highway. In this animal model study, the results indicated that PM10 had more respiratory effects while ultrafine had greater cardiovascular effects. These effects were observed irrespective of distance from the roadway; hence, at least in this study, it appears that toxicity may be driven more by particle size than a balance of fresh/aged components on the PM.

While the biological mechanisms and particle characteristics that influence toxicity of UFP are not well understood, there is enough evidence from animal studies to suggest that they are at least as toxic as PM2.5. While both PM10 and PM2.5 appear to have a toxicological effect, UFP might have a greater effect because of their relatively large surface area and their ability to be transported into the circulatory system. Regarding the smaller-sized UFP, it is unknown what proportion of UFP is in the size cut below 23 nm; therefore, control strategies that exclude this subset of UFP may not be health protective. Finally, studying the health effects of UFP using an epidemiologic approach remains problematic, due to the current lack of an adequate exposure assessment. However, as exposure assessment methods improve, we should gain a greater understanding of the extent of health effects attributable to UFP.

### 2.2 Proposed modifications to the current limit for PM mass emissions and the new optional solid particle number limit

It is proposed that beginning in 2014, all vehicles subject to LEV III-SULEV requirements must comply with at least one of the following two standards. The manufacturer can select either standard to demonstrate compliance.

- **Federal Test Procedure (FTP)-weighted PM mass emission limit to 0.006 g/mi in 2014 and to 0.003 g/mi in 2017**
  - PM mass emissions are measured over the FTP driving cycle using filter collection in a Constant Volume Sampling (CVS) dilution system followed by gravimetric weighing of the filter. The procedures are described in 40 CFR Part 86, and 40 CFR Part 1065. Adherence to relevant sections (defined in a future ARB test method document) of Part 1065 is required.

This emission limit is based primarily on new data currently being collected in a round robin exercise between ARB, U.S. EPA, and the car manufacturers. However, data found in the published literature and from previous ARB testing results were also considered.

- FTP-weighted SPN emission limit to $6.0 \times 10^{12}$ particles/mi in 2014 and to $3.0 \times 10^{12}$ particles/mi in 2017
  - SPN emissions are measured over the FTP driving cycle using CVS dilution and integration of real-time measurements. Measurements are obtained post-2.5 µm cyclone classification and using commercially available condensation particle counting instrumentation, hot dilution of sample, sample thermal treatment of volatile particles, and other requirements (i.e., conditioning and calibrations) that adhere to and comply with the new European requirements under the Euro 5/6 directives. The anticipated sampling approach to be required in the new California regulation is patterned after the Particulate Measurement Programme (PMP) measurement protocol, which became the basis for the new European regulation that mandates compliance with a SPN limit.
  - ARB staff is seeking public comment and specific suggestions for modification and improvement of the PMP approach. The procedures and calculations used (defined in a future test method document) are analogous to those of 40 CFR Part 86 Subpart B for integration of continuous gas measurements. Adherence to Part 1065 is required as appropriate for the elements of CVS dilution that overlap the mass measurement (i.e., pre-cyclone sampling requirements applicable to CVS system).

The stringency of the proposed amendments to the existing PM mass standard is meant to arrest the potential increase in new vehicle PM emissions expected due to the wider penetration of highly efficient GDI gasoline engines that are likely to be deployed for their CO$_2$-reduction benefits. Figure 1 illustrates a scenario for the penetration of GDI technology based on federal estimates. This simple projection does not incorporate any analysis of automaker-specific product plans for particular types of GDI technology deployment. Instead, the projection is a fit to the actual and estimated future populations for a fleet that complies with the 2012-2016 federal standards based on U.S. EPA data. It is acknowledged that the ultimate, post-2016 GDI share of the new light-duty fleet illustrated here is uncertain. However, ARB staff believes that the assumed GDI technology deployment shown in Figure 1 is a reasonable approximation for the trend in future light-duty engines. As more forecast data and detail on automakers’ particular product plans become available, staff expects to refine these estimations.

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12 Regulation (EC) No 715/2007, “on type approval of motor vehicles with respect to emissions from light passenger and commercial vehicles (Euro 5 and 6) and on access to vehicle repair and maintenance information,” 2007
**Figure 1.** GDI fleet penetration scenario based on fit to current and estimates for compliance with federal requirements.

It is expected that a variety of available and emerging technologies will be deployed to maintain PM emissions well below the LEV II emission standard of 0.010 g/mi and achieve SULEV certification. Generally, emission-control technologies found in current vehicles include close-coupled TWC, heated oxygen sensors, sequential fuel injection, and exhaust gas recirculation. For GDI engines, the technology for injecting and guiding the gasoline spray can have a significant impact on engine-out PM emissions. Based on ARB staff and industry-submitted data, gasoline injection systems with catalytic converter systems can achieve PM emissions below the proposed LEV III standards without necessitating the use of a gasoline particle filter (GPF). In addition, staff has received input from a number of manufacturers suggesting that a PM mass standard of 0.003 g/mi can be met by their future GDI engines without requiring the use of a GPF.

The proposed optional SPN standard is also meant to ensure that new vehicle fleet PM emissions do not gradually increase due to the increasing share of GDI or other low-\text{CO}_2 technologies. The new SPN standard is proposed at a level so that a GDI vehicle meeting the PM mass standard would also likely meet the SPN standard, and vice versa. The optional SPN limit is advantageous for a number of reasons, including increased industry flexibility, a stronger connection to health science on particle emissions, and the potential for improved measurement precision.

**Flexibility for compliance**

First, given the international nature of the car industry, a California SPN standard adds new and cost-saving flexibility for compliance. This comes from the opportunity for the car makers to make use of the same testing efforts they need now for compliance with the new European SPN limit. The intent is that by aligning this optional California
approach with the European regulation, the burden on the manufacturers due to the need for expanded testing is minimized.

**Update to the science underpinning the LEV program**

Second, over the last decade, an increasing amount of scientific evidence has emerged on the potential health effects of exposure to particles, both volatile and solid particles and in particular those in the ultrafine size range (< 100 nm). As discussed above, epidemiological studies show associations between ambient concentrations of PM mass and various health endpoint measures such as premature mortality or hospital admissions. There are very few similar studies establishing links between total UFPs and health endpoints, and there are no similar studies attempting to link SPN emissions to health endpoints. Nevertheless, the existing epidemiological and animal studies conducted thus far provide some evidence indicating that exposure to ultrafine particles may have adverse health consequences. The associations between particle exposure and adverse health impacts is particularly striking in urban areas and near roadways dominated by the emissions from motor vehicles - gasoline and diesel. The proposed regulatory change brings into focus the importance of considering the number of particles, not just their mass.

**Practical and superior measurement**

Third, one of the key lessons learned from the European effort that resulted in the SPN standard is that as PM emissions decrease, counting particles becomes a superior method to the gravimetric approach in terms of precision. It is also simpler and provides a test result more quickly. ARB tests conducted on DPF-equipped diesel vehicles confirm the higher precision of the particle counting method. Given the added precision, the option of counting particles to show compliance is believed to offer greater power to manufacturers for fine-tuning their technology designs.

**2.2.1 Counting the number of solid particles in vehicle emissions**

In this proposed regulation, SPN refers to the total number of particles emitted per mile that remain solid at temperatures of 300 °C, and that are greater than approximately 23 nm in diameter.

Particulate matter in vehicle exhaust refers to particles ranging in diameter from about 500 nm to 5 nm and smaller. These particles consist of a mixture of solid and semi-volatile materials. Solid particles are those which remain in the solid phase at high temperature. Semi-volatile particles are those which may be solid under some sampling conditions, but evaporate at high temperature or high dilution. The total number of particles measured using CVS dilution can be very sensitive to the measurement conditions, especially dilution - both the rate and the amount - and temperature. This sensitivity is caused by particle dynamics and the processes of condensation, evaporation, and nucleation of semi-volatile materials in the particle size range of 5 to 20 nm. In contrast, the number of solid particles is not sensitive to the sampling
conditions because they are not evaporating or condensing and the particle dynamics can be frozen by the measurement approach. For vehicular exhaust without a particle filter, the majority of the mass is emitted in particle sizes ranging from about 50 nm to 200 nm, and a large fraction of that is solid carbonaceous material (soot). To achieve a reliable particle measurement that is not sensitive to sampling conditions, which may vary from laboratory to laboratory, the PMP measurement protocol specifically excludes particles <23nm and particles that evaporate at or below 300 °C. ARB staff proposes to follow that approach and is seeking public comment on potential modifications that may improve the specific application of PMP to gasoline PM measurements.

ARB staff fully recognizes that Europe had a very different reason for adopting a particle number limit. In Europe, the new number standard is a technology-forcing step to promote wide adoption of particle filtration, which a PM mass standard alone at the level chosen could not guarantee. In California, staff is proposing to take advantage of the significant advances in metrology accomplished by PMP, and to utilize those new methods for the progress of the State’s regulatory actions for motor vehicles.

The filter sampling and gravimetric procedures described in 40 CFR Part 1065 were designed specifically in anticipation of the low PM mass emissions from DPF-equipped diesel engines. The modifications to the previous measurement approach (40 CFR Part 86) were necessary to verify compliance with an engine emission standard of 0.010 g/bhp-hr. Today, those protocols are fairly well understood and widely implemented for clean diesel measurements. The procedures, born out of unprecedented collaboration between industry, government, academia, and others, have been used by various laboratories to measure PM emissions with good single-laboratory precision from gasoline LDVs having emission rates as low as 0.001 g/mile. Emissions below this level begin to near the limit of detection of the CVS measurement. In addition, the Part 1065 methods require dedicated test facilities, infrastructure upgrades, and are time and resource intensive. In contrast, the SPN measurement is accomplished with commercially available instruments that were introduced to market to meet the need created by the standards based on the PMP protocol. Results can be available immediately after the end of a test; the resulting simplicity and speed allow the opportunity for much more emission testing to be conducted in support of vehicle development, compliance monitoring, and research.

Controlling the number of solid particles implies control of soot emissions because, in general, soot particles are mostly larger than 23 nm and are mostly solid. However, this approach does not provide control of sub-23 nm particles (semi-volatile or otherwise). The proposal to use the PMP method for measurement of SPN emissions in this regulation does not imply that ARB considers particles in the sub-23nm size range unimportant. In fact, the same can be said about the new phase of PMP work in Europe. In the international work exploring the application of PMP to heavy-duty engines commenced in 2008, there is now wider recognition of the importance of counting particles smaller than 23 nm, which may indeed be solid in nature. PMP attributes the
impetus for this work to findings by ARB\textsuperscript{13}. The PMP method is selected because it is the only particle emission measurement method that has undergone extensive international scrutiny and widespread inter-laboratory testing and verification. Unfortunately, the only exception is the United States, where the method has received serious attention only in California. As a result, the body of data to support conclusions regarding method performance and emission rates for future California GDI vehicles over California driving cycles is limited. ARB continues to explore the PMP measurement, opportunities for improvement, and impacts of particles outside the capabilities of the PMP method. Staff encourages public comment on the technical aspects of PMP, especially those related to the on-going Heavy-duty PMP Programme as they may relate to the proposed LEV III developments.

There are two approaches to reducing the number of solid particles in the emissions: incorporate aftertreatments such as particle filters, or improve the combustion process. In the diesel application, it is very difficult to eliminate soot emissions through combustion strategies alone – with the exception perhaps of low-temperature combustion. Therefore, in Europe, as discussed previously, the SPN standard (6 × 10^{11} km^{-1} over the NEDC) has been set to force use of DPFs. These standards become effective at the Euro 5/6 stage for all categories of diesel vehicles (M, N1, N2). The Euro 5/6 number standards must be met in addition to the PM mass emission limits. However, the Euro 5/6 PM mass emission limit is comparable to the LEV II limit, and is not a constraining factor on vehicles that use a particulate filter. Europe plans to establish an SPN standard for gasoline vehicles by September 1, 2014. It is possible that the standard will be set comparable to the standard for diesels, and thereby will force the use of GPF on gasoline vehicles.

Gasoline direct injection vehicles produce much less soot emissions than non-DFP diesel vehicles, but much more soot than gasoline PFI vehicles. Figure 2 is a collection of data found in the published literature and from ARB tests. The figure shows typical ranges in the number emissions for various classes of vehicles and driving cycles. Careful control of fuel/air mixing, and enhancement of the catalytic converter system can achieve very low emissions of particles. Although a GPF could achieve even lower particle emissions, ARB staff believes that the added cost, complexity, and magnitude of reductions from what are already very low emissions do not support that approach. In addition, the purpose of the proposed SPN standard is to limit backslide from the current PFI performance, not to establish a new tier of emission performance. Thus, ARB proposes to set the SPN standard at a level so that GPF is not needed.

Figure 2. Solid particle number emission rates for various types of vehicles and over various driving cycles. Data source is the published literature and ARB test results. NEDC is the New European Driving Cycle.

ARB staff acknowledges that additional study is needed and hopes to conduct future efforts with stakeholder participation. One important data gap is comparison of driving cycles. There are extensive data sets comparing SPN and PM mass emissions collected during the PMP method development and verification. However, these data are collected over the NEDC, and there are little to no data available at this point to develop robust relationships or conversion factors from NEDC to FTP emission rates. Also, most of the test data are for DPF-equipped diesel vehicles. Because DPFs remove solid PM, the relationship between PM particle number and mass will be different for post-DPF emissions than for GDI emissions. ARB has data and comparisons of NEDC and FTP emissions for a single Euro 4 compliant DPF-equipped LDV. The vehicle, referred to as the Golden Vehicle, was the reference standard for the 12 international laboratories, including California, that participated in the PMP inter-laboratory correlation exercises for LDVs. To begin addressing the data gap, ARB has recently measured PM particle number and mass emission rates over the FTP and UC driving cycles for GDI vehicles. ARB and auto manufacturers are currently testing additional vehicles. ARB results for six wall-guided GDI vehicles over the FTP are presented in Table 1. Data from four of these vehicles includes results for SPN emissions. Table 1 and Figure 3 show the results for individual phases of the FTP. Table 2 and Figure 4 show the FTP-weighted results. The regression line in both figures suggests that a total SPN limit of $3 \times 10^{12}$ particles per mile is similar to a PM mass limit of 0.003 g/mile. The regression lines in both cases show strong $R^2$.

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correlation, but the regressions are dominated by cold-start emissions (the four highest points in Figure 3), which are well above the proposed standard. Figure 5 is a magnification of Figure 3. It shows the expected scatter present at the very low emission levels that exclude the cold-start. It is noted that these hot-running and warm-start emissions are well below the proposed limits and their divergence from the regression line is not entirely surprising. The reduction of SPN concentrations will result in the reduction of soot, although it is acknowledged that a measurement-based one-to-one correspondence with the PM mass standard is not exact, and would be masked by experimental uncertainty of the filter-based method in any event. When the emissions consist primarily of soot (i.e. black carbon), there is a moderate correspondence between the number of solid particles and the total PM mass in the emissions. As the emissions become dominated by organic, volatile species, lack of correspondence is expected due to the inherent uncertainty in the experimental measurement and the fact that, fundamentally, each measurement is a different operational definition.

The proposal to regulate the number of particles emitted by a motor vehicle is new to California. It introduces new areas where additional thought and investigation are needed and staff is seeking public input. Some of the policy-relevant questions prompted by the new SPN regulation concept are related to the existence of particles below the PMP limit of 23 nm. First, metal-based additives in gasoline and lubricating oil in the absence of a particle filter have been shown to generate emissions of solid particles in the sub-23 nm size range. This raises the question as to whether the PMP protocol should be augmented by lowering the specified particle size-cut. This is the same question being entertained by the Heavy-duty PMP Programme in a group of new considerations for potential program improvements. Second, vehicle exhaust contains semi-volatile particles. These include organic compounds such as polycyclic aromatic hydrocarbons that are known to be harmful, and inorganic compounds such as sulfates whose health effects are uncertain. This raises a second question as to the need or desire to modify the PMP protocol in order to count all particles, not just the solid fraction. The public is invited to provide specific comment on these questions, especially and in particular those members of the public who may also be stakeholders in the European PMP process.
Table 1. PM Mass and SPN emissions for six wall-guided GDI vehicles over individual phases of the FTP.

<table>
<thead>
<tr>
<th>Veh</th>
<th>Cycle</th>
<th>Phase 1</th>
<th>PM mg/mi</th>
<th>SPN</th>
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Note: SPN emissions here are based on the original PMP protocol, which uses dilution factor for calculation of emissions. The most recent PMP method requires using a particle concentration reduction factor (PCRF), which accounts for particle losses. The difference between the two methods is about 25 percent higher SPN emissions when using the PCRF.

Figure 3. PM Mass and SPN emissions for six wall-guided GDI vehicles over individual phases of the FTP.
Figure 4. FTP-weighted PM mass and SPN emissions for four wall-guided GDI vehicles.

Table 2. FTP-weighted PM mass and SPN emissions for wall-guided GDI vehicles.

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<th>SPN</th>
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<td>1</td>
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<td>5.95E+12</td>
</tr>
<tr>
<td>2</td>
<td>5.97</td>
<td>4.39E+12</td>
</tr>
<tr>
<td>3</td>
<td>3.80</td>
<td>3.07E+12</td>
</tr>
<tr>
<td>4</td>
<td>3.79</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>3.29</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>3.07</td>
<td>3.98E+12</td>
</tr>
</tbody>
</table>
3. Mitigation of black carbon emissions for their air quality and global warming impacts

Along with California’s strides toward cleaner air, the State has a commitment to reduce its GHG emissions in order to contribute to efforts around the world to mitigate the effects of the accumulation of heat-trapping gases in the atmosphere that spur global warming. Since enactment of California’s Global Warming Solutions Act of 2006, the State has developed its “Climate Change Scoping Plan,” the general framework describing the mix of regulatory strategies, voluntary programs, and incentives designed to achieve the target GHG reductions. The Plan’s near-term goal is to roll back GHG emissions to 1990 levels by 2020 or about a 30 percent reduction from business-as-usual. The long-term goal is to reduce California GHG emissions 80 percent below 1990 levels by 2050, a goal that is consistent with international targets for long-term climate stabilization.

LDVs are substantial contributors to California’s GHG emissions, accounting for approximately 28 percent of the state’s total GHG emissions. As a result, the Scoping Plan states that a transformation of the LDV fleet toward lower GHG emissions is critical to achieving California’s climate change goals. Accordingly, the Scoping Plan includes

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a series of programs for increased vehicle efficiency, low carbon fuels, lower rolling
resistance tires, vehicle purchasing incentives, and electrification of the vehicle fleet. 
Approximately 18 percent of the total targeted 2020 GHG emissions reductions are
expected to come from the LEV III-GHG emission standards.

LEV III-GHG regulations build upon the original California Pavley standards that were
adopted in 2004 and the just-adopted U.S. EPA and NHTSA national program that
includes regulations for LDV GHG emissions through model year 2016. The existing
California and U.S. EPA GHG standards regulate carbon dioxide (CO$_2$), methane (CH$_4$),
nitrous oxide (N$_2$O), and hydrofluorocarbons (e.g., HFC-134a) from motor vehicles. This
section discusses the possible inclusion of BC in California’s rulemaking for GHG
standards. Setting a BC LEV III emission standard recognizes mitigation of PM
emissions for their air quality and climate impacts.

3.1 Black carbon as a climate warmer

PM is a regulated criteria pollutant of concern for the LEV III-SULEV standards. BC is
the light-absorbing carbonaceous fraction of PM that results from incomplete
combustion of fossil fuels and biomass. The heightened interest in BC mitigation today
is built on the well-recognized association of these emissions with localized air pollution
and their severe negative health impacts that are discussed above. Any climate
strategy for reducing BC emissions offers these important co-benefits.

When emitted into the atmosphere and deposited on ice or snow, BC converts visible
light energy to heat and warms the atmosphere. On snow and ice surfaces it reduces
albedo and causes melting, which generates a negative feedback loop of progressively
declining snow and ice surface area. Black carbon is also a particularly acute warming
agent. According to Jacobson (2010)$^{17}$, each gram of ambient BC in fossil-fuel soot
warms the air 1.1 to 2.4 million times more than each equivalent gram of ambient CO$_2$
and about 50,000 times more than each gram of ambient methane (CH$_4$). This acute
warming of the atmosphere and reduction in snow-ice albedo result in rapid increases in
global temperature and other impacts.

The average lifetime of BC is about a week, so the duration of its climate impact is
relatively short-lived. As a consequence, the reduction and elimination of sustained BC
emissions have the potential to provide a rapid near-term reduction in warming that
complements and supports climate change mitigation. Prominent scientists have
argued that large-scale BC emission reductions have the potential to delay the onset of
dangerous climate change and avoid rapid increases in atmospheric warming. In
addition, certain short-term and rapidly developing climate impacts such as the loss of
Arctic summer sea ice or alpine glaciers may be addressed most effectively by targeting
short-lived forcing agents like BC.

$^{17}$ Jacobson, M.Z., Short-term effects of controlling fossil-fuel soot, biofuel soot and gases, and methane on climate,
The Intergovernmental Panel on Climate Change (IPCC) has addressed the question of BC in its three previous assessment reports. In its most recent report published in 2007, the IPCC quantified the global radiative forcing of BC in the atmosphere and on ice and snow surfaces. Based on IPCC estimates, the cumulative impact of BC is equivalent to the third largest warming effect after carbon dioxide and methane. More recent measurement and modeling studies published in the peer-reviewed literature suggest this warming is underestimated by the IPCC. Ramanathan and Carmichael (2008) recognize BC as a significant contributor to climate change, second only to CO\textsubscript{2}\textsuperscript{18}.

Black carbon was also a topic at the ARB’s Haagen-Smit Symposium in 2009. The symposium is an annual event to foster discussion and interaction among policy makers, researchers, and the regulated community. This last symposium focused on addressing the missing pieces of California’s carbon footprint\textsuperscript{19} and how some climate-active pollutants and their emission sources are not fully integrated into current policy. Challenges and co-benefits associated with BC emissions reductions were noted. A number of issues that may impede policy action include the fact that BC emissions are not covered in the Kyoto Protocol and the co-emission of BC with cooling pollutants, namely organic carbon (OC), complicates accounting and development of effective interventions. However, the symposium discussants clearly recognized the enormous potential health co-benefits due to reduced PM2.5 exposures.

Estimates of the carbon-equivalent emissions of GHG emissions use a weighting factor known as the Global Warming Potential (GWP), which is the ratio of the cumulative radiative forcing due to the instantaneous release of a given mass of pollutant over a time horizon (typically 100 years) relative to the same mass emission of CO\textsubscript{2} over the same time horizon. Thus, the GWP is a measure of the relative effectiveness of a warming agent to cause a change in temperature relative to CO\textsubscript{2}. A GWP is needed in order to estimate the carbon-equivalent emissions of a pollutant in GHG emission inventories and allow for a simple comparison of expected impacts.

While the IPCC did not publish a GWP estimate for BC in its most recent report, independent estimates have been published in the peer-reviewed literature, including estimates drawn from IPCC report itself. These suggest that BC is capable of generating warming that is two orders of magnitude greater than carbon dioxide. On a 20-year horizon, which places greater emphasis on rapid, near-term climate impacts, this BC warming is three orders of magnitude greater than the CO\textsubscript{2} warming (see Table 3). Still the exact value to use when quantifying the BC impact continues to be debated, including the extent to which the location and time scale of warming should be addressed. Staff is seeking specific input not only on how to address these issues, but also on how to select an appropriate GWP value for BC. Staff is also aware of the debate over alternative metrics to the GWP, but these are not being strongly considered.


given the absence of international dialogue and the predominant role that the GWP plays in international policy. Nevertheless, staff invites input on how alternative metrics could be incorporated into the LEV III proposal and coupled with state and international climate policy.

Table 3. Black carbon GWP from recent studies shown for two time-horizons.

<table>
<thead>
<tr>
<th>Source</th>
<th>Black carbon global warming potential</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>100-yr</td>
</tr>
<tr>
<td>Bond and Sun, 2005(^{20})</td>
<td>680</td>
</tr>
<tr>
<td>Hansen et al, 2007(^{21})</td>
<td>~500</td>
</tr>
<tr>
<td>Reddy and Boucher, 2007(^{22})</td>
<td>480</td>
</tr>
<tr>
<td>Forster et al, 2007(^{23})</td>
<td>510</td>
</tr>
<tr>
<td>Fuglestvedt et al., in press(^{24})</td>
<td>460</td>
</tr>
<tr>
<td>Rypdal et al., in press(^{25})</td>
<td>920</td>
</tr>
</tbody>
</table>

The choice of time horizon for the GWP value that is used to estimate the statewide BC inventory and to be incorporated in the ARB staff proposal may have significant consequences. Table 4 shows a simple approximation of LDV carbon-equivalent emissions using a 20-year and 100-year GWP for all regulated GHG pollutants (CO\(_2\), CH\(_4\), N\(_2\)O, and HFC) from an average vehicle today and a future vehicle. This shows that short-lived pollutants like BC and CH\(_4\) are expected to contribute a relatively small fraction of the net warming of a vehicle. The two time frames can lead to a four-fold difference in CO\(_2\)eq emissions. Table 4 presents the product of the GWP and GHG and BC emissions. Future BC emissions are based on 0.003 g/mi PM emissions and an assumption of 66 percent BC fraction in PM. 500 GWP\(_{100\text{-yr}}\); 2000 GWP\(_{20\text{-yr}}\) are used. A manufacturer may choose to simply equate the BC emission rate to that of the proposed PM mass limit in lieu of measuring BC exhaust emissions.

ARB has not created an official inventory of BC emissions in California and one is needed. However, ARB’s emissions inventory for criteria pollutants and the U.S. National Emissions Inventory include PM emissions, which provide a basis for building a bottom-up BC and OC emissions inventory. BC and OC emissions can be estimated


from source-specific PM2.5 emissions and the relative BC and OC fractions in the emitted PM. Research has already yielded relevant information. According to an ARB-sponsored study by the Desert Research Institute\textsuperscript{26}, the major source of BC emissions in 2006 in California was wildfires (29 percent of total BC emission). BC emissions from on-road mobile sources accounted for 20 percent of the total and these are dominated by diesel exhaust. The study also quantified the emissions of OC. As discussed previously, there are concerns about the relative increase in future years of the gasoline LDV contribution as low-CO\textsubscript{2} technologies such as GDI gain fleet share. Preliminary source testing of PM components between the GDI and non-GDI vehicle indicate that, depending on the component, the emission rate from the GDI vehicle is a factor of 2 to 8 that from a PFI vehicle, and BC highly exceeds other PM chemical components.

Table 4. Approximate illustration of equivalent CO\textsubscript{2} emissions from different GHGs for 2009 and for future (2020 and beyond) year new vehicles.

<table>
<thead>
<tr>
<th>GHG emission</th>
<th>Global warming potential\textsuperscript{a}</th>
<th>100-year gCO\textsubscript{2}e/mile</th>
<th>20-year gCO\textsubscript{2}e/mile</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO\textsubscript{2}</td>
<td>1 1 337 &lt;200 337 &lt;200</td>
<td>377 0 17 17</td>
<td></td>
</tr>
<tr>
<td>AC CO\textsubscript{2} (indirect)</td>
<td>1 1 17 TDB 17 TBD</td>
<td>16 0 1.8 0.5</td>
<td></td>
</tr>
<tr>
<td>AC refrigerant</td>
<td>1430 3830 6 0</td>
<td>298 289 0.1 0.03</td>
<td></td>
</tr>
<tr>
<td>CH\textsubscript{4}</td>
<td>25 72 1.8 0.5</td>
<td>25 72 1.8 0.5</td>
<td></td>
</tr>
<tr>
<td>N\textsubscript{2}O</td>
<td>298 289 0.1 0.03</td>
<td>298 289 0.1 0.03</td>
<td></td>
</tr>
<tr>
<td>BC \textsuperscript{b}</td>
<td>500 2000 0.3 1.0</td>
<td>500 2000 0.3 1.0</td>
<td></td>
</tr>
</tbody>
</table>

\textsuperscript{a}From IPCC 2007 fourth assessment review (AR4) except BC global warming potential estimate is based on ARB review of scientific literature. Pavley I used IPCC 2001 TAR GWPs values (e.g. 1300 for HFC-134a, 23 for CH\textsubscript{4}, 296 for N\textsubscript{2}O). 100-yr GWPs are IPCC 2007 AR4 (1,430 for HFC-134a, 25 for CH\textsubscript{4}, 298 for N\textsubscript{2}O) as used by U.S. EPA. 20-yr GWPs are IPCC 2007 AR4 (3,830 for HFC-134a, 72 for CH\textsubscript{4}, 289 for N\textsubscript{2}O)

\textsuperscript{b}Based on 3 mg/mile PM and 0.66 BC/PM fraction

Controls on BC emissions from LDVs will invariably cause reductions in emissions of the non-BC fraction of PM, which consists predominantly of OC. This acts as a cooling agent when emitted in the atmosphere, reflecting visible light and offsetting the warming impact of BC. Therefore, the benefits to the climate from BC controls may be tempered by reductions in OC. For the purposes of comprehensive accounting, it would make logical sense to include OC in the GHG calculation for fleets. However, this proposal does not include cooling agents in a future GHG basket since this may generate unwanted tradeoffs to public health. Staff seeks input on this approach and how best to account for the OC cooling agent in the PM emissions.

3.2 Proposed new limit for BC emissions

There are a number of different possible approaches for inclusion of BC in GHG

standards applicable in 2017. Staff is primarily considering two approaches. The first is the CO₂-equivalency approach that was adopted as part of the existing California Pavley standards for model years 2009 and later vehicles, whereby all the GHG emissions are bundled in the same CO₂-equivalency framework. In order to put BC into a CO₂-equivalency framework, three components are required. These are the vehicle PM mass emissions, the BC fraction of those PM mass emissions, and the appropriate GWP for BC. ARB staff has not formulated an approach as to whether the BC fraction would be derived from actual vehicle emissions tests or a default value from existing information. In addition, further development is needed should a car maker choose to comply with the SPN limit instead of the PM mass limit and the use of that information for determination of BC emissions.

The second approach would include per-vehicle limits for BC that are not bundled into CO₂-equivalency standards. This approach is more similar to how the non-CO₂ emissions of N₂O and CH₄ are treated by the U.S. EPA for the national 2012-2016 GHG standards. These two approaches have very different data requirements and would represent different degrees of effort on the part of ARB and the regulated industry. A per-vehicle limit would potentially require additional testing. In contrast, the bundle approach may be more amenable to use of default values. Public input is sought as to the most viable approach for achieving the intended climate benefits.

3.3 Method for determination of BC emissions

The lack of an agreed-upon definition and the most appropriate emission measurement method are two important unanswered questions regarding determination of BC emissions from motor vehicles. One of the major open issues that requires attention is the development of a traceable standard for BC or EC since one will be needed for calibrations and assessment of instruments interferences. However, the recent heightened interest in BC is prompting progress in these areas. As discussed below, measurement methods have been established for years and are frequently used for testing ambient air monitoring and vehicle emissions. In addition, there are several emerging instrument options that could prove useful when integrated into new measurement methods. Thus, ARB staff is confident that, with public input, agreement upon a measurement method is feasible in the timeframe of this rulemaking.

The nomenclature often used in referring to carbon emissions is illustrated in Figure 6. Black carbon particle matter emissions from incomplete combustion processes are often referred to as soot, which is composed of carbon and other byproducts of combustion. Black carbon is often used interchangeably with soot, but more recently has been operationally defined as those PM emissions that are quantified in the exhaust using light attenuation techniques. Elemental carbon (EC) and OC contribute large fractions to fine particle mass in motor vehicle emissions. Elemental carbon is determined as the mass on a PM filter that can be ascribed to carbon based on its tendency to pyrolyze under controlled conditions after the OC fraction has been volatilized. Studies have shown that the BC and EC methods correlate reasonably well.
The most common methods for determining BC and EC are filter-collection methods, including optical and thermal methods. In optical methods, such as the aethalometer, aerosol particles are first deposited on a filter and the increase in light attenuation through the filter is measured and is proportional to an equivalence mass of BC. In thermal methods\textsuperscript{27}, such as IMPROVE\_A and NIOSH 5040, the filter is heated and carbon is thermally evolved at controlled temperature steps and in controlled atmospheres (i.e., non-oxidizing vs. oxidizing). The result is an operational definition of the concentration of EC and OC. Different temperature and optical monitoring protocols result in different values of EC even within the same thermal evolution method. The separation of OC from EC is ambiguous because some of the EC combusts in the presence of oxygen, and some of the OC can turn to EC in an oxygen-deficient atmosphere.

- **Elemental Carbon:**
  - Mass determined based on operational defined analytical method
  - A few different quantification methods, such as IMPROVE\_A
  - Used for emission inventories

- **Soot:**
  - Impure carbon emitted as the results of incomplete combustion
  - A description of incomplete combustion

- **Black carbon:**
  - Light absorbing carbon.
  - A description of physical (light) property of carbon
  - Global Warming Potential

**Figure 6.** Comparison of definitions commonly used when referring to the solid fraction of PM.

The filter-based thermal method offers several advantages for the measurement of BC. There is a large database available nationwide for emission inventory as well as for assessing visibility impact (Regional Haze Rule). ARB staff believes that this mass-based quantification method can be immediately used for assessment of carbon-equivalent impacts in conjunction with an appropriate GWP. The method enjoys wide use for assessment of health impacts, emission inventories, and ambient air monitoring in the State Implementation Plan. Furthermore, recent study by Chow et al (2009) developed carbon analysis quality assurance and quality control procedures

including: (1) multi-point temperature calibrations; (2) characterization of analysis atmosphere; (3) carbon analyzer calibration; and (4) calibration of laser intensity using neutral density filters. These procedures have been shown to improve the precision of OC/EC and carbon fraction measurements.

In principle, the relatively strong light absorption of BC can be used to infer BC from an optical measurement of aerosol light absorption and knowledge of the mass specific absorption of BC. This approach has the advantage, from a climate perspective, that the primary measurement relates directly to the light-absorbing properties of the aerosol. Commercial instruments commonly used based on optical methods such as the particle soot absorption photometer, the aethalometer, or the multi-angle absorption photometer measure an attenuation signal that is related to the absorption coefficient of the sampled mass of aerosol. The photoacoustic analyzer, which directly measures the absorption coefficient, detects the acoustic signal produced when the sample stream is heated via the absorption of laser light by particles in the air sample. These instruments have the advantage of providing continuous or semi-continuous results, allowing monitoring of transient events. However, conversion of the measured light-absorption to mass relies on an empirically-determined conversion factor, which may change with the source.

In summary, the relatively short atmospheric residence time of BC makes reductions in BC emissions a potential near-term opportunity to postpone the effects of rising GHG levels on the global climate. Control measures to reduce BC climate-forcing emissions often would have collateral benefits by also reducing emissions of health-related pollutant. A better characterization of mobile source emissions of climate forcing and air pollutants will help improve the understanding of co-benefits of future emission reduction strategies. The public is invited to provide specific comment on these subjects.