

The authors are grateful to the referee 2, Mr. Nikiforos Zacharof, for giving us the critical insights and suggestions. The manuscript has been carefully modified based on the opinions. Sometimes there were difficult parts to be modified and the authors explained why we did not make change. The modified sentences based on the referee 2's suggestions are highlighted in green.

## *1 General comments*

### *1.1 Introduction*

*The authors should pay more attention to the introduction and make clear what is the purpose of the paper. There are several statements about the representativeness of the laboratory testing compared to the on-road measurements, but it is not clear why this is discussed, while both approaches have pros and cons. Without proper listing of them cannot enable a comparison between the two approaches. In this section, it is needed to clarify better the goals of the paper and why the authors conducted this research.*

Thank you for the suggestion. According to the opinion, the sentence was modified to promote the 'purpose' of this study to be easily understandable for the readers. The final goal of this study has already been included in the final sentence of Introduction.

Lines 75-78: "The purpose of this study is two-fold. First, chassis-dynamometer-based and RDE measurements using PEMS were conducted on a heavy-duty Japanese vehicle to determine the importance of RDE specific factors, including the ambient temperature and road gradient, among others. Second, the obtained experimental results were analysed based on two parameters, i.e., the driving force and vehicle speed, ..."

### *1.2 Methodology*

*Please describe the methodological approach (testing, analyses, etc) by offering an overview before moving into describing the testing process. With this structure, the reader is confused as the text moves directly into explaining the testing, but there has been no frame to put it into. Also, a clear description of the post-processing and the analyses that are performed to reach the goal of the paper is missing. Please provide an overview of the approach, describe each step of the approach and indicate all the analyses that you've performed. Additionally, please avoid presenting equations that a reader would already be familiar to, e.g. formula of acceleration.*

The authors convinced for the opinion and following sentences were added in the revised manuscript. The redundant formula in equation (2),  $dv/dt$ , was removed according to the suggestion.

Lines 85-89: "Three methods were used in this study: laboratory test using the chassis dynamometer, RDE measurements using PEMS, and data analysis to construct the estimation model for the emissions inventory. For a detailed analysis of the experimental results from the RDE measurements, data processing methods included data smoothing for the high-resolution time profiles of the emissions data and the extraction of the road gradient from official open sources. Details on these methods are described in sections 2.1 to 2.3."

### 1.3 Results and discussion

*The authors need to improve the presentation of the results in terms of text structure and language. Several times it has been unclear what they wanted to say, while some other times the text was repetitive. Please present all the analyses that help you answer those questions that you have set. At this point, it seems only a simple presentation of the measurement results. This is useful to enhance the literature in the topic, but it does not add up significantly, while there is potential for this. Please consider adding a more detailed sensitivity analysis, an energy analysis (energy share on wheel, auxiliaries, etc) and an improved comparison between the WHVC and on-road conditions. These are just recommendations to improve the paper, based on what is already included. Consider expanding the analyses as you see fit.*

Thank you for the suggestions. The structures of the discussion were completely revised based on the three reviewer's opinions. The authors treated sensitivity analysis section 3.4.2 as the complement explanation to the transient table analysis to clarify what factors are important to the exhaust emissions. Hence, if it is possible, we do not want to further discuss the sensitivity topic in this manuscript (further, enough measurement data to be used in the sensitivity analysis in the suggestion were not collected). The next PEMS experiment is now ongoing, and the authors are intending to take detail sensitivity analysis including energy share or the effect of vehicle parts into consideration. Again, we are grateful for the valuable advices.

### 1.4 Conclusions

*The conclusion sector is unclear to what outcome the analysis reached. This section should avoid repeating what has already been presented in the discussion and actually reach to some conclusions and evaluate whether the paper's goals have been achieved or not, what new questions have arisen and what could be done next. The sole outcome is that the road gradient is the most important factor affecting NOx and CO2 emissions, which is hardly new. The authors conducted an experimental campaign that should gain visibility, but it needs more analyses, reach some solid conclusions that could actually contribute in the field.*

Conclusion sometimes repeats the outcome described in the previous sections to highlight the important topics in this study, especially base experimental results and transient table analysis. The finding of the effect of road gradient is not new but quite important in this study, so the authors decided to retain it. The novelty of this study was added in the Introduction as the following sentence.

Lines 80-81: "To the best of our knowledge, this is the first study in which PEMS measurement results are applied in the development of an estimation model for vehicular exhaust.

Based on the suggestion, to make clear the final goal of this research with what has still not been unsolved, following sentences were added in the revised manuscript."

Lines 327-331: "The experiment as well as the detailed analysis were conducted only for one heavy-duty vehicle in the Japanese market. In future, it is expected that further studies would be

conducted to obtain the variability of real-world vehicular exhaust emissions; after the measurements results for the consistent number of vehicles have been obtained, the emissions inventory based on real-world measurements should be evaluated for use in policy making regarding air quality treatments.”

## *2 Specific comments*

*Line 20 Please provide references and/or examples of major countries/regions that are concerned with pollutants.*

According to the suggestion, related references were added with following sentence.

Lines 22-23: “...including the United States, the European Union, China, India, and Japan (Akimoto et al., 2015; Costa et al., 2014; Ravindra et al., 2016; Sullivan et al., 2018; Yang and Wang, 2017).”

*Line 23 Please provide reference to the NASA report or database.*

Thank you for the clarification. The exact reference has been provided by the webpage as following.

National Aeronautics and Space Administration Webpage.: <https://climate.nasa.gov/vital-signs/global-temperature/> (accessed on December 15, 2020).

*Line 25 The Saito 2010 reference is already 10 years old. Please add also a projection from newer studies if possible.*

The authors are convinced about the opinion. Following recent reference from IPCC (2018) was added in the manuscript.

Summary for Policymakers. In: Global Warming of 1.5°C. An IPCC Special Report on the impacts of global warming of 1.5°C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty. [https://www.ipcc.ch/site/assets/uploads/sites/2/2019/05/SR15\\_SPM\\_version\\_report\\_LR.pdf](https://www.ipcc.ch/site/assets/uploads/sites/2/2019/05/SR15_SPM_version_report_LR.pdf) (accessed on November 24, 2020).

*Line 25 – 26 “... photochemical oxidant is a well known short-term air pollutant ...” It is not clear which photochemical oxidant the authors refer to and also there’s lack of any reference to back this statement. Please elaborate on the whole sentence.*

Thank you for pointing out the lack of information. The following sentence was added with the citation of Sillman (1999).

Line 29: “...photochemical oxidants, mainly composed of ozone,...”

Sillman S. The relation between ozone, NO<sub>x</sub> and hydrocarbons in urban and polluted rural environments. Atmos. Environ., 33, 1821-1845, [https://doi.org/10.1016/S1352-2310\(98\)00345-8](https://doi.org/10.1016/S1352-2310(98)00345-8),

1999.

*Line 30 – 31 “To address the problems of global warming and photochemical pollutants, it is necessary to mitigate air pollution.” This statement is not quite accurate. Pollutants cause health problems mainly and also sometimes have a global warming potential. The major factor for global warming from the automotive sector are CO2 emissions. CO2 is not a pollutant and it doesn't cause health problems by inhaling it. Please elaborate.*

Thank you for pointing out the ambiguous expression. CO2 is not a health-hazardous component but despite this fact, CO2 is categorized as long-term air 'pollutant' when it is emitted by the anthropogenic sources such as vehicles, power plants etc[1]. Therefore, the sentence was modified as following.

*Line 35: “...to mitigate the emissions of long- and short-term air pollutants.”*

[1]<https://www.nationalgeographic.com/environment/global-warming/pollution/#:~:text=Though%20many%20living%20things%20emit,as%20gasoline%20and%20natural%20gas.>

*Line 40-41 “In general, the laboratory temperature is set at approximately 25 °C, and it cannot be easily changed via the normal laboratory system.” This is not quite true, the temperature in the laboratory and more specifically in the vehicle test cell can be adjusted to a range of different temperatures. In some cases, however, it could require additional investment in infrastructure such as to achieve a temperature of -7 °C that are required in some countries. The 25 °C temperature is mandated by the testing protocol and the regulation that has adopted this protocol and not necessarily from the technical capabilities of the laboratory. Please elaborate the sentence.*

Thank you for the detail clarification. The previous sentences indicated the difficulty of arranging arbitrary room temperature because of the exhaust heat from the laboratory system including the heat from vehicular exhaust. The sentences were modified as following.

*Lines 45-46: “...it cannot be easily set to the arbitrary temperature (especially a low temperature range) owing to the exhaust heat from vehicle and measurement machines via the normal laboratory system...”*

*Line 43 “... a specific activation temperature that cannot be attained in cold seasons ...”, this is quite a bold statement and not entirely true. The activation temperature will be reached at some point, but under cold conditions, it could take longer. In the case of small trips and low temperatures, then it is possible that the activation temperature is not reached. Please elaborate.*

Thank you for the clarification, this sentence was also pointed out by another reviewer. The relationship of ambient temp. and pollutant's emission is described in the results section, so in the introduction, the sentence was modified as following.

Lines 47-50: “It has been noted that environmental temperature considerably influences the amount of exhaust emissions (detailed explanations regarding this observation are provided in section 3.1), leading to the release of a large amount of pollutants (including NO<sub>x</sub>) into the atmosphere in low ambient temperature conditions (the MOVES2010 Report by the U.S. Environmental Protection Agency, 2010).”

*Line 44 – 46 “Moreover, the road gradient also influences the amount of exhaust emissions because it directly affects the driving force, which is presumed to be proportional to CO<sub>2</sub> and other exhaust emissions.” This statement is not correct. The gradient increases the required engine load and it indeed increases CO<sub>2</sub> emissions, but this is not done necessarily linearly. The road gradient force formula is defined as  $m \cdot g \cdot \sin(a)$  ( $m$  = vehicle mass,  $g$  = acceleration of gravity,  $a$  = road grade). The engine needs to operate at a higher load to compensate for this force, but the operation point depends also on the transmission ratio. In any case, the road grade increases fuel consumption and therefore CO<sub>2</sub> emissions – not necessarily linearly. In the case of pollutants, they could increase but this depends on the engine operation point that affects what kind of pollutants are produced in the engine. Whether these pollutants make it to the exhaust relies heavily on the operation of the aftertreatment systems.*

Thank you for the clarification. The authors also agree that road gradient does not linearly affect to the driving force, and it depends on the parameters of engine torque and transmission ratio. In the analysis of this study on Fig.4 and Fig.5, the emission tables were evaluated treating with the two variables: driving force and vehicle speed. Vehicle speed was chosen to exhibit the engine rotation factor which corresponds to the transmission ratio, for example, in the high transmission range, engine rotation would increase. After-treatment systems introduced in the tested vehicle were EGR and urea-SCR. Fig.4 and Fig.5 treated those two after-treatment contribution by separating the effect of after-treatment tools in terms of operation time of the tools. Meanwhile, the term ‘proportional’ was modified by “...have a negative effect on...”.

*Line 46 “Consequently ...” This statement does not exactly explain why the governments are making PEMS tests and it seems like a leap of thought. Also no references are provided to back this statement. The following statement in line 48 “Gallus et al. (2017) ...” refers to an experimental campaign and not to the adoption of PEMS as an official protocol to define emissions. The following references also refer to research campaigns and not protocols. Please re-phrase and elaborate.*

According to the suggestion, the following sentence with several references related to the political issues of RDE was added.

Lines 54-59: “For this reason, the European Union has implemented a regulation for RDE from light-duty vehicles using portable emissions measurement systems (PEMS) (Valverde et al., 2020). Consequently, countries, such as the United States, China, and India, have made decisions to implement RDE measurements as a regulatory test, and Japan is now considering the implementation of regulations via real-world measurements using PEMS (Giechaskiel et al., 2019).

Currently, in terms of the research field of atmospheric science, many researchers have conducted RDE measurements using PEMS.”

*Line 55 “Nevertheless, the conduction of road measurement experiments using PEMS is a relatively new domain, and only a few studies have been performed to assess the analytical data ...” This is not quite true, please check the literature on this issue an elaborate. PEMS is common in testing, especially on light-duty vehicles, where in the European Union at least is part of the vehicle type approval procedure for pollutants requires Real Driving Emission (RDE) testing (Regulation (EU) 2017/1151). The cold start is important for light-duty vehicles as they are often doing short trips and the aftertreatment systems do not reach always optimal operation temperature. However, for heavy-duty vehicles the cold start effect is limited as the vehicles operate for long time (e.g. 8 hours or longer for a typical city bus operation) and they are compensated by the long operation time.*

Thank you for the clarification. RDE by PEMS has been adopted as the type approval test in 2017, which is relatively (or quite) newer than chassis dynamometer test. We added “compared with chassis dynamometer experiments” in lines 67-68.

The driving duration of heavy-duty vehicles depends on the use of the vehicle. City bus could be driven continuously for a long duration but the vehicle for house-move could be sometimes driven with stop and run. The emission from short-time parking is sometimes equivalent to the cold-start emission especially in the cold season. Further, heavy-duty vehicles will not be substituted by the zero-emission vehicles in the near future because of the difficulty of high capacity battery. For those reasons, the RDE experiment for heavy-duty vehicle is also important to mitigate both current and future air quality issues.

*Line 60 The term “classical mechanics” is a bit redundant in this context. Please consider to elaborate.*

According to the suggestion, the words “classical mechanics” were modified as a following sentence.

Lines 77-78: “Second, the obtained experimental results were analysed based on two parameters, i.e., the driving force and vehicle speed, to develop...”

*Line 62 Please take into consideration the instrument accuracy. PEMS can be utilized everywhere on-road, but could face accuracy issues, while these problems are minimized in the laboratory where the methodology and the instruments (e.g. bag result analysis) could be more accurate.*

According to the suggestion, the sentence regarding the accuracy of PEMS was added with the citation of the study from Cao et al (2016).

Lines 65-69: “The accuracy of PEMS is also important in discussing RDE test results. Cao et al. (2016) conducted a study to clarify the accuracy of gaseous pollutants measured by PEMS, concluding that NO<sub>x</sub> emissions would sometimes be overestimated in the low NO<sub>x</sub> concentration range, accounting for a ~50 % maximum from the reference NO<sub>x</sub> concentration owing to analyser

drift. These results suggest the limitation of PEMS accuracy when the measurement system is used in the type approval test.”

*Line 98 The Eq. 1 seems redundant, but it could be retained. However, what the authors are describing here is a rolling average with a step of 5 observations. Please clarify.*

Thank you for the suggestion. The detail explanation of eq.1 was added after eq.1 as the following sentences.

Lines 129-133: “In equation (1), vehicle speed was smoothed using the 5-point average of the speeds at neighbouring time steps. We determined the averaging number of vehicle speed as 5 points based on two concepts. First, the averaging number of vehicle speed should be minimized as much as possible to maintain the high resolution time step. Second, the dispersion should be sufficiently lower than the measured vehicle speed. It is also worth noting that the averaging number of vehicle speed may depend on the measurement tools, and the 5-point value was suitable in this study.”

*Line 101 The Eq. 2 seems redundant as it presents the calculation of acceleration based on speed, which must be a well-known topic for the reader. Retain it if you consider it useful for your narration, otherwise please remove and elaborate.*

The authors thought that eq.2 is important to show the concrete definition of central differential method although it is a well-known mathematical method. But we also thought that the definition of acceleration,  $a=dv/dt$ , is actually redundant and this definition was deleted from eq.2.

*Line 103 The authors in this section (2.3.2) describe the synchronization of the data. For every data that are retrieved from the vehicle there's a time lag between the phenomenon that occurs and the measurement time. Combustion process and exhaust emissions are directly correlated as the latter are products of the former process but the data are retrieved with a time lag as gases need to travel from the combustion chamber to the exhaust where they are measured. Data timestamps from the combustion chamber and the exhaust would correspond to different events and they need to be synchronized. This applies to all the sensors but not all the events can be correlated in this way. This section seems that it is not needed as this process is standard for the post-processing. However, if you retain it please elaborate this section and consider merging it with another one where you describe the data post-processing.*

Actually the data synchronization is required for vehicle exhaust measurements when the detail analysis is conducted. The synchronization method could be different among the researchers or engineers, and in this study, the optimization by the cross-correlation function was chosen. It is important to show the definition of synchronization method since the method is not widely known by the researchers or engineers in this field. The sentences were retained but the section 2.3.2 was merged to 2.3.1 based on the suggestion.

*Line 120 The  $\mu_r$  and  $\mu_a$  are not defined. Please define that are the rolling resistance and air drag coefficients.*

Those two parameters were defined as “rotation friction coefficient” and “air friction coefficient” in lines 141-142, so the authors would like you to check it again.

*Line 122 You mentioned the vehicle weight was set at 5880. Have you measured the vehicle on a balance or did you derive this value from your calculations?*

Vehicle and PEMS weights are catalog value. Battery weights were measured. The following explanations were added in the manuscript.

Lines 156-158: “The total vehicle weight,  $m$ , including the weight of the vehicle itself (4,920 kg), cargo, such as PEMS ( $\approx 200$  kg), four batteries for PEMS ( $37 \text{ kg} \times 4 \approx 150$  kg), driver and operator ( $55 \text{ kg} \times 2 \approx 110$  kg), and other measurement-related parts (500 kg), was set as 5880 kg”

*Line 142 The “engine room” could mean the whole compartment where the engine is placed. Please consider replacing it with the more appropriate term “combustion chamber”.*

According to the suggestion, “engine room” was replaced by “combustion chamber”.

*Line 186 “which is not taken into account by the WHVC approach.”. The WHVC development was based on real-world data in order to produce a representative situation of real-world conditions. The claim that the testing conditions are not represented in the WHVC is useful as observation, but it needs to back it with enough data. First, it should be quantified how many times are encountered the testing conditions that were outside the WHVC approach. The following questions must be answered. Has it been on every test, has it been affected by the driving style or has it been due to the requirements of your experiment? Second, it is needed to compare to real-world route conditions to the overall WHCV approach and quantify the effect and its significance.*

Thank you for the clarifications. The authors were convinced about the opinions and following sentences were added in the revised manuscript.

Line 227: “...were observed in all RDE test conditions.”

Lines 228-231: “We note that the acceleration distribution also depends on the driving feature, and the distribution changes for each driver (Ericsson, 2001). The drivers of the RDE and chassis dynamometer experiments were different in this study, such that the results of the difference in the acceleration distribution can also be attributed to the difference in the driving feature.”

As described in the manuscript, the route of RDE in this study did not include high speed range which is included in WHVC. For this reason, the authors did not compare the detail of PEMS results and WHVC results. PEMS measurement with high-speed range in the motor-way RDE measurements are now ongoing, and the results will be opened in the coming year. Thank you for your reconsideration.

*Line 199-207 This is methodology, please move to the respective part.*

Thank you for the suggestion. After the careful consideration based on the suggestion, we thought that it is better to retain the sentences at this part. The method to obtain transient map from driving force, vehicle speed, and NO<sub>x</sub> and CO<sub>2</sub> emissions has already been described in the Methodology section. On the other hand, the background of the transient map is described at section 3.4.1. The background could also be described in Introduction, but we thought that it is more reader-friendly to remain these sentences (Lines 243-247) at the top of section 3.4.1. Please reconsider our decision.

*Line 217 - 218 It should be clarified that the use of air conditioning poses an additional load to the engine that could make the engine operate in less optimum operation ranges under some conditions. In this way it could lead to an increase in pollutants.*

Thank you for the suggestion. Based on the suggestion following sentence was added in the revised manuscript.

Lines 264-265: “The use of the air conditioner leads to an additional load on the engine that may render engine operation at less optimum operational ranges, leading to an increase in emissions.”

*Line 245 – 250 It is stated several times the that “a parameter depends on the same parameter” such as “the road gradient depends on the cosine of the road gradient”, which is quite obvious. Please elaborate the whole text and remove such statements.*

Thank you for the clarification. The term was modified from “road gradient” to “road angle”. It seems there is no similar miss-sentence.

*Line 253 – 254 “the WHVC driving mode is currently applied worldwide.” I am not sure whether this is true and it is quite vague. Please mention major countries/regions that use this protocol and for what reasons. In Europe for instance, the heavy-duty vehicle type approval procedure is performed through a simulatory approach that utilizes other driving cycles.*

WHVC is mainly used for the research purposes to measure and compare exhaust emissions for various heavy-duty vehicles (although WHVC is also required for the imported vehicles from abroad which could not be tested by engine bench.). The type approval tests, on the other hand, is composed of engine bench tests, WHSC and WHTC, in Europe, North America, Japan, and Australia (<https://dieselnet.com/standards/cycles/index.php>). To account for these issues, following sentence was added in the revised manuscript.

Lines 309-310: “...including Europe, the United States, and Japan, among others, mainly for research purposes (DieselNet Webpage).”

### *3 Technical corrections*

*Line 28 “(O’Neill et al., 2004; Chappelka and Samuelson, 1998; Wang et al., 2017)” please re-arrange in*

*chronological order.*

Thank you for pointing out the order of citations. The order was modified chronologically.

*Line 44 Please provide an accurate reference for the EPA.*

The citation was modified to be “Report of MOVES2010 by U.S. Environmental Protection Agency, 2010”. The exact reference was described as following.

U.S. Environmental Protection Agency: MOVES2010: Highway Vehicle Temperature, Humidity, Air Conditioning, and Inspection and Maintenance Adjustments. [https://www.google.co.jp/url?sa=t&rct=j&q=&esrc=s&source=web&cd=1&ved=2ahUKEwjZrOKxvOzoAhWB3mEKHSJWCf0QFjAAegQIARAB&url=https%3A%2F%2Fpub.epa.gov%2Fsi%2Fsi\\_public\\_file\\_download.cfm%3Fp\\_download\\_id%3D504585%26Lab%3DOTAQ&usg=AOvVaw1oHKO1-hMuLXe2RqyY5868](https://www.google.co.jp/url?sa=t&rct=j&q=&esrc=s&source=web&cd=1&ved=2ahUKEwjZrOKxvOzoAhWB3mEKHSJWCf0QFjAAegQIARAB&url=https%3A%2F%2Fpub.epa.gov%2Fsi%2Fsi_public_file_download.cfm%3Fp_download_id%3D504585%26Lab%3DOTAQ&usg=AOvVaw1oHKO1-hMuLXe2RqyY5868) (accessed on January 16, 2020).

*Line 54 “(Kousoulidou et al, 2013; Kwon et al, 2017; Liu et al, 2009; Luján et al, 2018; O’Driscoll et al, 2016)” please re-arrange in chronological order.*

Thank you for pointing out the order of citations. The order was modified chronologically.

*Line 70 “current Japanese regulation set in 2016.” Please cite the exact law. Line 76 “... in our previous work...” Please avoid using possessive pronouns and especially in the first person. Consider removing them entirely and retain the reference to your work. If you want to retain the connection with your work for any reason, please consider using the third person e.g. “in the authors’ previous work”.*

The current regulation for heavy-duty vehicle was added in Table S1 of supplementary information. The possessive expression of the citation was removed to “in the previous study”. Thank you for the clarification.

*Line 259 “in the Japanese market were conducted”, I think you mean a vehicle that is available in the Japanese market, but it unclear. Please correct.*

The term ‘used’ was added before the words “in the Japanese market were conducted”.