

Author response to referee comments

Atmos. Meas. Tech. Discuss., amt-2017-305

We would like to thank the two referees for their detailed comments and suggestions to improve the manuscript. We hope that the re-submitted manuscript addresses all concerns at this stage.

Title: HIGH SPATIOTEMPORAL RESOLUTION POLLUTANT MEASUREMENTS OF ON-BOARD VEHICLE EMISSIONS USING ULTRA-FAST RESPONSE GAS ANALYZERS

Notes about the organisation of this document:

The comments of the two referees (R1 and R3, respectively) have been addressed sequentially, with changes signified by page and line number related to the original submission.

In the case of similar or overlapping content in the comments (e.g. a general comment is satisfied by the resolution of a specific comment, or if one response/action addresses multiple comments), we give the full details of the change only at the location of the comment which required the most significant changes. All other related comments make reference to the item with the full response, identified by comment number. The full response may be earlier or later in the document than the comment in question.

Black text (hereafter):

Referee comment [with additional surrounding text for necessary context given in square brackets]

Blue text:

Response: Our comments to the reviewers and the public

Action: What we have done to change this in the text of the article

From Referee 1:

Comment: I recommend publication after minor changes, although I feel that the paper could gain further scientific relevance with a more in-depth data analysis of the driving experiments.

Although the authors wish to “keep the focus on the technique and the instrumentation” (page 3, line 6), a better data analysis will lead to more interesting conclusions or recommendations (e.g. for reducing traffic emissions by changing driving styles or adapting urban infrastructure), and would give more direction to follow-up research.

Response: We’d like to thank the reviewer for this comment. A number of changes have been made throughout the manuscript (see below) in an attempt to

give more direction to follow-up research using similar techniques, where the focus would be on real world emissions and not the technique and instrumentation (e.g. a model and measurement sensitivity study of different routes incorporating differing levels of urban and rural infrastructure would be very informative to policy makers).

Action: P7: Section 4.2 – ‘Further Work’ added, replacing P7, L18
“Further improvements have already been made to the spatial accuracy of the positioning data. Real-time Kinematic (RTK) GPS has been successfully implemented allowing an accuracy of <1 cm to be achieved in favourable conditions, with an increased 15 Hz logging frequency. This has further increased the benefit of the spatial emissions positioning of this technique.

This paper discusses the specific technique required to take high spatiotemporal emissions data but consideration of the implications of such data is important. GPS plots of emissions hot spots could provide important information for local air quality councils and transport planning authorities. However due to the relative set-up times of such testing it is unlikely to be a useful tool for in-service compliance checks. If used correctly, emissions maps could provide recommendations on changes to improve local air quality. The most obvious change, from a purely scientific point of view, would be the removal of all speed bumps as these cause transients that contribute significantly to high NO_x levels. Their placement close to schools or pedestrian areas reinforces this argument due to the direct impact on local air quality. Such a change would however have other political and safety implications far beyond the scope of this paper. Further, improvements through synchronisation of traffic lights in order to streamline traffic flows may be possible. A greater uptake in driverless or at least intelligent vehicles that could feedback to a centralised traffic control centre would aid this. Drivers could also shoulder some responsibility for emissions improvement so education in better driving techniques or even the inclusion of a driving quality metric into licencing tests could be beneficial. Although this study includes several tests on two differing vehicles, the data set is insufficient to allow the above hypothesis to be tested. Further work in repetition of specific road elements in different vehicles, and employing different driving styles, is needed.”

Comment: In Section 2 (page 4, line 5) it is explained that the emissions from the gasoline car are sampled “pre-muffler but post 3-way catalyst”, which is in conflict with the description in Section 2.2 (page 5, line 1-3) which state that NO is sampled both upstream and downstream of the catalyst. In the latter case: can the authors present some numbers about their measured efficiency of the catalyst? Is this as expected? How does it react under transient driving conditions?

Response: To clarify, for this vehicle, there are two sampling points; one before the catalyst, and one after the catalyst but before the muffler. In this study, the analyzer measured from both sample locations, however, only post-catalyst data has been published. The text has been appropriately changed.

On gasoline vehicles, catalyst efficiency is usually measured to be 100% at all steady state conditions once catalyst conversion temperature has been reached. Tailpipe

emissions breakthrough is measured where transient pedal input upsets catalyst conditioning (reductants), and NO_x is no longer completely converted.

Action: P4 L5 Text changed “(...) fitted through the vehicle floor to two sampling points, one before the three-way catalyst and one post catalyst but pre-muffler. For engine calibration applications, having pre and post-catalyst sampling is essential to understanding aftertreatment operation and excellent time alignment. For air quality applications, analyzing post-catalyst data is more beneficial. The diesel data {...}”

Comment: Page 2, line 11: abbreviation RDE is used, while only explained at page 5 (line 19) for the first time.

Response: Agreed, this should be explained at first use.

Action: P2 L11 Text changed to define RDE at point of first use: “(...) of a Real World Driving Emissions (RDE) test (...)”

Comment: Page 4, line 13-15: maybe it should be remarked that the diesel vehicle is not equipped with a catalyst.

Response: Yes, this would provide extra context to the reasons for emissions breakthrough of this vehicle.

Action: P4 L15-6 Text added: “The vehicle was in good repair and in current use with no form of NO_x aftertreatment (e.g. a catalytic convertor). For fleet context (...)”

Comment: Page 4, line 32-33: “(...) as it was anticipated that the NO_x emissions would be relatively low (NO_x being mainly a byproduct of diesel-powered internal combustion engines)”. I presume that the authors mean NO₂ instead of NO_x (and that therefore there was no need to use an NO₂ converter).

Response: Yes, this is a mistake and will be changed.

Action: P4 L32-3 Text changed, NO_x to NO₂: “as it was anticipated that the NO₂ emissions would be relatively low (NO₂ being mainly a byproduct of diesel-powered internal combustion engines)”

Comment: Page 5, line 27: “Figure 3 shows (...) increasing from 20 mg/s baseline when stationary”. From the figure I would estimate 2 instead of 20 mg/s.

Response: Agreed, text has been changed in the document.

Action: P5 L27 Text changed to “Figure 3 shows (...) increasing from 2 mg/s baseline when stationary”

Comment: Page 5, line 27-32: The authors are measuring both NO and NO_x, so I would expect some words about their findings in the NO/NO_x ratio. Is the NO₂/NO_x ratio approximately stable (also during the acceleration phases)? If so, is it relevant to keep measuring NO₂ in future experiments?

Response: The mechanisms by which NO and NO₂ are created are different. For a gasoline vehicle, very little NO₂ is produced during combustion, therefore, measuring NO only is sufficient. The ratio of NO₂:NO_x in a diesel vehicle varies during transients up to approximately 30% dependent on air-fuel ratio, engine speed, load amongst other factors. It is therefore important to measure both NO and NO_x because the addition of the NO_x converter allows the most dangerous pollutant, NO₂, to be calculated from the difference; interesting for air quality purposes as NO₂ is individually legislated. Whilst NO_x-only may be sufficient for atmospheric research; NO₂ is often desirable for Air Quality research.

Action: No publishable data at this time.

Comment: Section 3.2, page 6, line 9-11: “Using much slower conventional PEMS equipment (...), this highly time-resolved event would be significantly delayed, and smoothed out over a longer period making its location difficult to place.” I would also remark that the duration is so short (~2s) that its magnitude would be missed by PEMS, having a response time of about 1 second.

Response: Very true, this is a key advantage of fast-response equipment. Having a fast enough analyzer ensures that the full magnitude of each emissions peak can be fully resolved.

Action: P6 L10-11 Text changed to: “(...) this highly time-resolved event would be significantly delayed, and smoothed out over a longer period. The true magnitude of this event would be missed due to its short duration and its spatial location would be difficult to place.

Comment: Section 3.2, page 6, line 12-13: To put things into perspective, I would include a remark that the current Euro emission standards are 60 mg/km (petrol) and 80 mg/km (diesel).

Response: Agreed, this will provide extra meaning to the data, however, the current standards are not necessarily relevant due to the age of these vehicles, so have provided current and calibrated values for both.

Action: P6 L9 – Text added: “{...} emissions peak (for reference, the current emissions standards – Euro 6 at time of publication – are 60 mg/km NO_x for Gasoline and 80 mg/km Diesel) {...}”

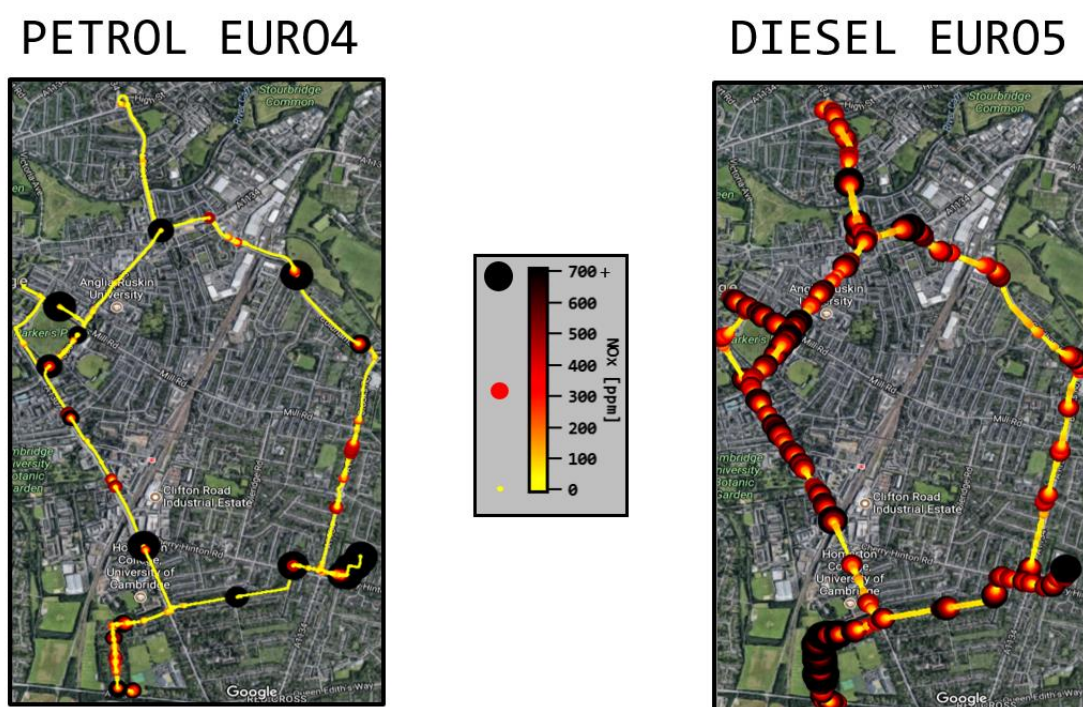
Comment: Section 3.3, page 6, line 19-20: From Figure 2b I learn that also the petrol car has been driven over these speed bumps. Why not include a comparison between the petrol and diesel vehicle emissions regarding speed bumps, as more or

less promised on page 4, line 26: “(...) used for comparison of diesel and gasoline vehicle emissions.”?

Response: We agree that this is a good comparison, and one that we have made. However, we would rather avoid starting a Diesel vs Petrol debate since the paper is focussed on the measurement technique.

Action: P4 L25-6 Text also added based on previous reviewer comments: “The GPS data for the London drive was set to 1 Hz (due to an earlier version of acquisition software), and the drive duration was 2.5 hours for the 53 km of this route (shown on the left in Figure 1). An urban route around Cambridge UK passing near continuous air quality monitoring stations was also designed and used for comparison of diesel and gasoline vehicle emissions (shown on the right in Figure 1).”

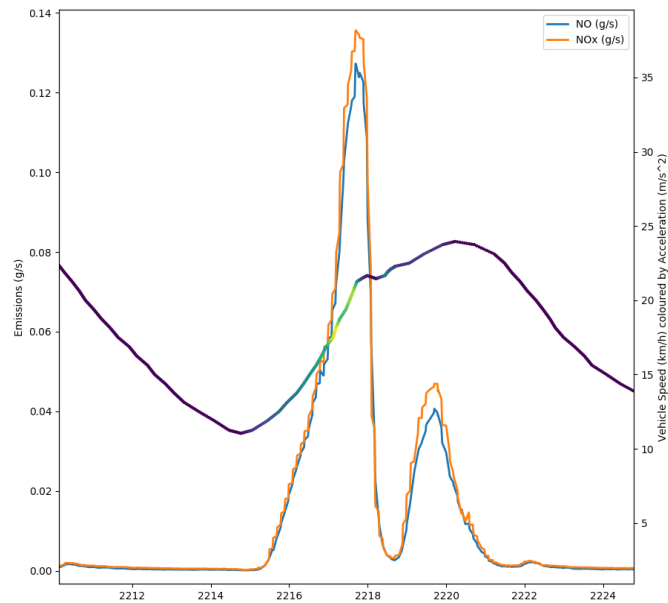
We can also provide the referee with some example data directly:



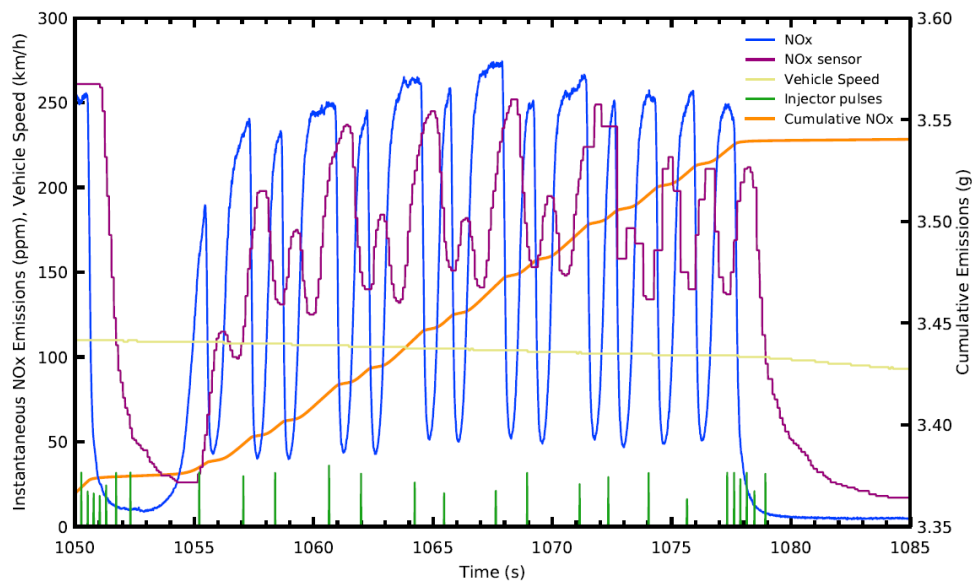
Comment: Page 6, line 30: “illustrating that acceleration (...) alone is not a suitable proxy for emissions.”. Why not? If I look at Figure 6a, I see two different velocity gradients (roughly 2216.0-2217.5s and 2218.5-2220.0s), corresponding to different emission peaks. If the sampling rate of the speed permits, it would be insightful to overplot this graph with the vehicle’s acceleration.

Response: It is true that in this case, where the vehicle is not fitted with any after treatment system (catalyst, SCR, etc) the acceleration is a good proxy for the ppm emissions (as seen in the first figure below), however this will fall down quickly for any modern vehicle with after treatment. This can be seen in the following plots, a Diesel fitted with SCR at steady state (and slight deceleration) can produce tailpipe

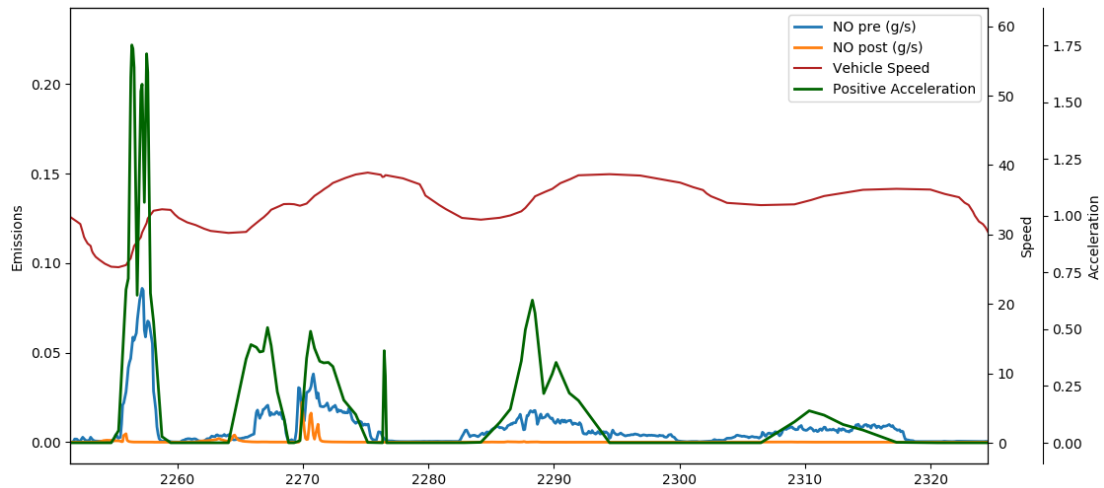
NO_x (confidential data), and an example from the gasoline vehicle included this paper, fitted with a catalyst, where breakthrough occurs only when the catalyst is poorly conditioned (pre-catalyst in blue, post in orange).



Euro 5 Diesel emissions with vehicle speed, coloured by acceleration.



Euro 6 Diesel, fitted with SCR. The SCR complicates the correlation.



Action: P6 L30 Remove text: “illustrating that acceleration (gradient of the velocity profile) alone is not a suitable proxy for emissions.”

Comment: Section 4, page 7, line 4-5: “the analyser’s sampling rate of 100 Hz captures emission transients that would be lost or smeared when using conventional PEMS equipment or other slower analysers”. I would include this important statement also in the abstract.

Response: We think this is a good idea, the abstract has been edited to include this.

Action: P1 L5 Text changed to: ‘The fast response time of the analyzers, which results in 100Hz data, makes accurate time-alignment with the vehicle’s Engine Control Unit (ECU) signals possible. This enables correlation with transient air fuel ratio, engine speed, load, and other engine parameters, which helps to explain the causes of the emissions “spikes” that Portable Emissions Measurement Systems (PEMS) and conventional slow response analyzers would miss or smooth out due to mixing within their sampling systems.

Comment: Figure 1: Consider to include arrow heads on the dotted lines to indicate direction of data flow.

Response: Another good idea.

Action: P10, Figure 1 has been edited to include arrows indicating data/electricity flow.

Comment: Figure 3b: Missing legend/color bar. It would also be interesting to see the location of all traffic lights along the driving path.

Response: The colour bar had been mistakenly left off this figure – the adjusted figure includes this.

Action: P12, Figure 3 has been updated to include a legend and colour bar.

From Referee 3:

Comment: I found that the introduction of the paper lacked a bit of context. The authors should sketch who they think would be interested in these measurements, and why. Can they be applied by national emission agencies for better emissions estimates, or are they useful for inspection authorities checking whether vehicles comply with EU-standards? Such information is missing from the paper. For illustration: the introduction now ends with what is “beyond the scope of this paper”, but it would be better to formulate clearly what will be addressed in the paper. Also, I strongly support the suggestion by Referee #1 who calls for a link of the observations to Euro emission standards for good perspective.

Response: The referee makes a good point. The introduction has been changed to include typical uses of the data and to outline the contents. Added emissions standards as per Reviewer 1’s request (see above).

Action: P2 L27 – Text added: “One of the applications of this technique is to help resolve emissions calibration issues on-board a vehicle. Portable {...}”

P3 L3 – Text added: The technique which will be discussed in this paper is the instrumentation of ultra-fast gas analysers for tailpipe sampling and the identification of urban road features conducive to producing high emissions for multiple vehicles. In addition, this data could be used by city councils or planning authorities to improve current road layouts or influence future developments to improve urban air quality.

Comment: Another concern I have is on the accuracy and the precision of the ultimate data provided (g/s and g/km emitted). The authors did not make any statement on the uncertainties in measuring the NO_x concentrations at the tailpipe. This should be repaired, and I also think an uncertainty estimate should be provided for the emission strength estimates.

Response: A full error propagation has been conducted for the data taken for this study using the variance method^[1]. A section outlining the process and assumptions has been added to the discussion. Where values have been stated in the text, uncertainties have now been provided. The calculations by which these uncertainties have been made can be found below:

$$f = f(x, y, \dots); \quad \delta_f = \sqrt{\left(\frac{\partial f}{\partial x}\right)^2 \delta_x^2 + \left(\frac{\partial f}{\partial y}\right)^2 \delta_y^2 + \dots}$$

And so where the live grams were calculated via^[2]:

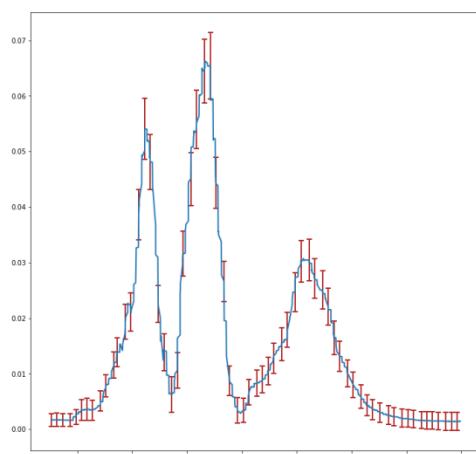
$$m_{\text{gas}} = u_{\text{gas}} \times c_{\text{gas}} (\text{ppm}) \times \frac{q_{\text{mew}} (\text{g/s})}{1000}; \quad q_{\text{mew}} = q_{\text{maw}} \times \left(1 + \frac{1}{A/F_{\text{st}} + \lambda}\right);$$
$$A/F_{\text{st}} = 14.7$$

Partial differentials with respect to c_{gas} , q_{maw} and λ with $\delta c_{\text{gas}} = \pm 50\text{ppm}$.

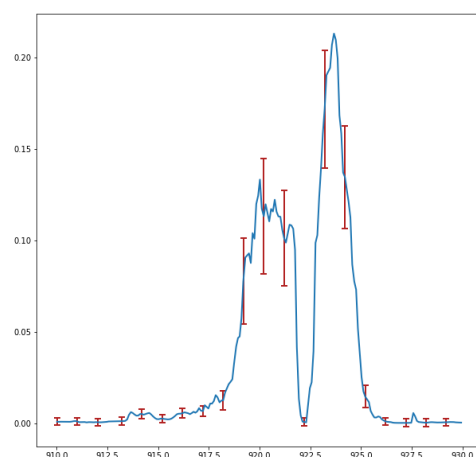
For the diesel (on board mass flow meter) an uncertainty value of 3% was used for δq_{maw} and this was calculated element by element. Lambda uncertainty of $\delta\lambda = \pm 0.5$ was used. The gasoline vehicle, equipped with a lambda sensor, has an uncertainty as shown in table below (calculated element by element) but also requires a volumetric efficiency calculation using an empirical comparison with a Bosch mass flow meter. A cross correlation was used to estimate the error on a scale every 10 kPa which was then used as a lookup element by element.

Accuracy at lambda 0.8	0.80 ± 0.01
Accuracy at lambda 1	1.016 ± 0.007
Accuracy at lambda 1.7	1.70 ± 0.05

Plots below for a section of diesel + section of gasoline:



1: Diesel, using on-board MAF and assumed lambda.



2: Gasoline, using on-board lambda and empirical MAF calculation.

[1]:

nistdigitalarchives.contentdm.oclc.org/cdm/compoundobject/collection/p16009coll6/id/99848/rec/1

[2]: <http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32016R0427&from=EN#page=45>

Action: P6 L34 – Section 3.4 added to describe uncertainty propagation.

“Section 3.4 – Error Propagation

An error propagation...”

P5 L27 Text changed to add uncertainty: “20 mg/s” changed to “ 2.0 ± 0.8 mg/s”

P5 L28 Text changed to add uncertainty: “70 mg/s” changed to “ 70 ± 6 mg/s”

P6 L9 Text changed to add uncertainty: “120 mg/s” changed to “ 120 ± 36 mg/s”

Comment: IF the authors wish to make the case that their method is superior to a conventional technique as PEMS, I think it would be good if they attempt to quantify how much better the detail is that they can detect now. Would that be relevant given the potential application of these data for e.g. vehicle compliance monitoring?

Response: In reality, the quantification of the detail benefit is difficult to calculate due to it being a composite of the spatial and the temporal resolution. Spatial resolution is 10x improved due to faster GPS logging and temporal resolution is improved by 100x due to the increased time response and logging rate. This has been included in the discussion.

Action: P7 L5 Text Added: “(...) or other slower analyzers. The ultra-fast analyzers therefore present a time resolution improvement of two orders of magnitude over PEMS. In addition, PEMS GPS data is recorded at 1Hz, whereas the location data in this study is logged at 10 Hz. This realises a 10x spatial resolution benefit in the logged location data and a 100x analyzer response time benefit, which matches the resolution of the GPS. The contribution of these aspects will give a vast spatiotemporal improvement.

For an application such as the identification of pollution “hot-spots” for the improvement of urban air quality, it is important to understand any road features which promote transient accelerator pedal input on a local scale, and therefore using a measurement technique able to sample many points throughout a short duration transient is fundamental to understanding its causes.

Comment: P2, L20: dependant → dependent

Response: Missed spelling/grammatical error.

Action: P2 L20 Text changed to “(...) largely temperature dependent (...)”

Comment: P3, L11: Bajaj et al (2002) should be entirely between brackets.

Response: Agreed, this has been changed.

Action: P3 L11 Text changed to “(...) inherent to GPS (Bajaj et al., 2002).”

Comment: P4, L22: “the deposition concentrations” is formulated a bit weird. With deposition, most atmospheric scientist understand the process of atmospheric constituents being deposited at the Earth’s surface. Here it appears to describe the constituents emitted to the atmosphere. Please clarify to avoid confusion.

Response: The wording could be made clearer. Text changed to avoid confusion.

Action: P4, L22 Text changed to “This sampling technique measures the tailpipe concentrations at street level”

Comment: P12, caption Figure 3: please also include in the caption that this figure holds for a diesel car.

Response: Agree with this change.

Action: P12, Figure 3: Text changed to “Time series of NO_x, NO, emissions in g/s of the diesel vehicle with vehicle speed of a traffic light pull-away in central London, UK. The red box represents the spatial component of the graph data, shown in the context of the city area.”