Air pollution near roads: An experimental and modelling study

Reply to reviewer 1

Nov 2017

Referee #1 Comments:

General comment. The analysis and discussion in this paper are not comprehensive. Overall, this paper is not well written. The authors tried to do so many things in this article, but there is no good story. There are lots of sections and sub-sections where the reader cannot get the full picture. Discussions are limited in many cases, and many conclusions are not well substantiated by their analysis results. It seems like the authors are trying to include several aspects in this paper, but there is no comprehensive view. The QA/QC of data is not well documented.

Reply:
We thanks comments from our reviewer and appreciate his/her effort to provide comments to improve our manuscript and our work.

Before replaying this general comment, we clarify that we use the following terminology regarding particles.

- *PM (Particulate matter): solid phase particles, regardless of their size, particle size distribution, morphology or chemical composition*
- *TSP (Total suspended particles or fine particles). Particles with aerodynamic diameter <30 μm,*
- *PM10: particles with aerodynamic diameter <10 μm,*
- *PM2.5: particles with aerodynamic diameter <2.5 μm,*
- *UFP: particles with diameters in the range of 1-100 nm.*

We oriented this manuscript towards the description of the temporal and spatial variations of traffic related pollutants near non- urban roadways by using a calibrated model which solve, via CFD, the equations that model the physics of dispersion of solid and gas phase species in a gas phase media under the varying conditions of the low troposphere (Pg 3, line 9). We highlight that:

- *We did not oriented this manuscript towards the description of the CFD model because: i.) it will make the manuscript too long, ii.) we believe that those contributions are not of the interest for the ACP audience. Then, we decided to include in this manuscript a brief description of the most relevant aspects of the model and its experimental validation. We also decided to fully describe the model in a companion paper, which is already under evaluation. (Pg 3, lines 10-13)*
- *We did not oriented our manuscript towards the description of the spatial variations of traffic related pollutants near roads based only in our experimental measurements because:*
  - *The purpose of the experimental work was to validate our NR-CFD model and then, use the calibrated NR-CFD model to study the effect of the varying conditions of traffic on near road pollutant concentration, in terms of short and long term concentrations, so that they can be used to asses human health impact.*

- *It will limit the validity of our conclusions to the specific conditions and timeframes under which we developed our experimental work (unpaved roads, 1 month of measurements, in a tropical area).*

Despite the contributions related to the implementation of the NR-CFD model are being published elsewhere, this manuscript reports the most important contribution of our overall work (pg 3, line 9).

- *We systematically used our NR-CFD model to determine concentration profiles downwind in the horizontal and vertical direction as function of meteorology parameters, emission rates and physical properties of the pollutant.*
- *We proposed non-dimensional number for pollutant concentration, distance from the road and showed that all gas-phase pollutants exhibit the same profile, and all solid phase pollutants exhibit the same profile.*
- *We developed a methodology to include the temporal variations into the analysis and to provide integrated long-term averages of concentration downwind, which are useful to evaluate human health impact of those pollutants.*
- *We measured near two unpaved roads, for long periods of time (~1 month), simultaneously, meteorological variables, traffic conditions, PM2.5, PM10 and TSP concentrations at 4 locations near the road, using instrumentation and protocols recommended by the USEPA and WMO.*

Manuscript was modified to emphasize on this orientation and the scope of our work. We also improved the description of the experimental work and results obtained from those measurements.

Major comments.

In this paper, the authors measured and modeled coarse (TSP, PM10, PM2.5) PM fractions downwind of two arterial roads. Many previous near-road studies have demonstrated that the traffic-emitted PM in a near-road setting is mostly dominated by ultrafine particles (< 100 nm), whereas coarse (TSP, PM10, PM2.5) PM is mainly dominated by regional/local background particles. The traffic-related pollutants (ultrafine particle, BC, NOx, CO, etc.) have strong near-road gradients, whereas near-road gradient of coarse PM is typically very mild (Karner et al., 2010). So one could have a strong concern that at which extent their measurements are relevant to traffic emissions? Although the authors performed their measurements at downwind of unpaved road, therefore, a large fraction of measured coarse PM are coming from the road dust. My concern is that how their measurements are relevant for a typical traffic emission/near-road perspective. For example, if someone wants to apply the knowledge from their paper in a typical near-road setting. Since the title of their paper says 'Air pollution near arterial roads'- thus, someone might expect the influence of traffic related pollutants (combustion pollutants; ultrafine particle, BC, NOx, CO, etc.) at first, not that much about coarse PM. I think the author should have a strong justification on how their measurements fit in a context of typical traffic-emissions/near-road environment. If the coarse PM is critical for a traffic/near-road perspective under particular environment, then the authors should reframe their paper, its title and analysis- center around coarse PM (since they only have coarse PM measurements) and that particular environment. As it is, to me, their measurements and analysis do not represent a typical near-road/traffic-related pollutants scenario.

Reply: *ok. Manuscript was modified.*

We understood that the main concern of the reviewer is that our measurements and modelling work included fine particles (0.1 <d <30 μm) but did not include UFP (d< 100 nm). Thus, our manuscript is not describing the dispersion of pollutants near a typical near road environment, which our reviewer consider to be the one near paved roads with high traffic of diesel and gasoline fueled vehicles.

We are looking for a larger scope than only paved roads and UFP. As stated previously, the focus of this manuscript is the description of the dispersion of pollutants (fine particles, UFP, non-reactive gases) near roads (paved and unpaved) by solving the differential equations that describe the physics of dispersion of gas and solid phase species in a gas phase media under the conditions of the low troposphere. Therefore:

- *We used the experimental work to calibrate this NR-CFD model for the case of fine particles*
- *We used the calibrated NR-CFD model and the experimental measurements to describe the spatial and variations of fine particles near unpaved roads.*
- *We extrapolated the use of our calibrated NR-CFD model to study the dispersion of gases near paved and unpaved roads. We found that results agree with reported experimental results (Chaney, Cryer, Nicholl, and Seakins, 2011).*
- *We highlight that pollutants near roads, besides tailpipe pollutants and pollutants present in the background, includes pollutants resulting from the interaction tire-road (see replay to comment No. 1). Thus, re-suspended road particles are also traffic related pollutants and they are also present in paved roads, independently if the vehicles moving on the road surface are powered by gasoline/diesel engines o electric motors.*

Then:

- *Attending reviewers concern, we are including a new section, where we again extrapolated the use of our NR-CFD model to the case of UFP for the case of the paved roads with high traffic of gasoline or diesel fueled vehicles. We also compared to experimental measurements reported in the literature and found good agreement. See description of UFP dispersion at the end of this document.*
- *We are highlighting in the manuscript that the physics of dispersion of UFP is different that of physics of dispersion of fine and coarse particles.*

Specific comments

Specific Comment 1. Background vs. roadway impact: How did the authors separate re-suspended PM from unpaved road vs. traffic emitted larger particles? This is important if exploring the influence of traffic emission is a primary goal of their study?

Reply:
We understood that reviewer refers to traffic emitted particles as those particles emitted from the vehicle exhaust tube and traffic emitted larger particles as background particles.

We clarify that near road pollutants includes:

- *Background pollutants, which are those pollutants originated from the surroundings, exhibit a constant concentration, and depend on the specific place of study. Thus, they become relevant when we are interpreting experimental data or when we are considering interactions between background pollutants and the ones originated from the source under study.*
- *Traffic related pollutants: Pollutants originated from the circulation of vehicles on the road. These pollutants can be classified according to their origin in:*
    - ***Tailpipe emissions:*** *These pollutants are originated from the combustion processes of engine-powered vehicles. They include ultrafine particles (UFP), CO, CO2, NOx, SOx, and unburned hydrocarbons, among which VOCs are of high interest. Those pollutants are of high interest, especially for the case due to their human impact. These pollutants are not present for the case of electric-powered vehicles.*
    - ***Tire-road interactions****: Emission of coarse (d >30 μm), fine (0.1 <d <30 μm) and ultrafine (d< 100 nm) particles result from the tire-road interaction due to tire wear, breaks wear, and resuspension of road particles. This source of particles is always present in both paved and unpaved roads.*

Then:

- *Traffic related emissions include both: tailpipe emissions and particles originated from the tire-road interactions. In the study of near road pollution, we should include both types of particles.*
- *In the case of unpaved roads, particle emissions due to resuspension are at least four orders of magnitude higher than tailpipe emissions. We identified that particles trapped in the high-vol filters came from resuspension and we could not identify tailpipe particles in the SEM analysis. This was due to the fact that tail pipe particles had a negligible concentration compared to the ones originated from resuspension.*
- *We did not intent to measure UFP in our experimental work.*
- *Background particles are different from re-suspended particles due to the interaction tire-road. In our experimental work, we selected two regions with negligible particle background concentration. We selected two regions fully covered with grass and no houses or any source of particles in a circle of at least 1 km of radius.*

In conclusion, we did not need to separate tailpipe particles from re-suspended particles to accomplish our objective of calibrate our NR-CFD model.

Specific Comment 2. Method section: there should be a clear description of what they measured, what instruments they used, how did they maintain QA/QC and data quality, instrument response time, data averaging time, sampling frequency, etc. These are very important given the near-road environments are very dynamic, in general. The detail on these can put in the supplementary. A table should be given summarizing all the important aspects related to instrumentations and data quality. There is no details about their sampling, variability, measurement uncertainty, etc. Did they measure continuously?

How many sample they collected at different locations and for how many days? There is no real mention (Fig) about their measured data and its variability. Also, based on their 24-hr filter sample, how did they tell anything about traffic influence since traffic is very dynamic? With their 24 hour filter sample, they essentially do not have any temporal information. For example, the influence of meteorology (boundary layer variation), traffic (diurnal traffic variation).

Reply: *ok. We are including additional details of the experimental measurements and data analysis. We are also including the measured data as supplementary material.*

As our driver is the assessment of near road air pollution on human health, we are interested in short (1, 8, 24 hr) and long (1 year) term averages of pollutant concentration downwind from the road to contrast them with air quality standards. The air quality standards are the max values of exposure (concentration during a given period of time) below which studies have shown to be safe for human health.

From the experimental perspective, the USEPA have stablished the recommended practices to determine pollutants concentration, which includes instrumentation technical characteristics and measurement protocols. Those protocols stablish the duration of individual measurements. For example, the USEPA have stablishes the determination of PM10 concentration through weight differences of filters exposed during 24 hr to a constant volumetric flow. We followed those protocols and determined PM10 and TSP concentration simultaneously in 4 points near the road.

The USEPA have also incorporated within its recommended practices the use nephelometer (light attenuation technique) for the determination of PM10 and PM2.5. This technique allows to have 1 min measurements, but for air quality assessment those measurements should be averaged for periods of 24 hrs. We incorporated two instruments that use this technique in our measurements. We measured every 10 min and reported 24 hrs-averaged values. (pg 5 line 10).

For the case of the meteorological variable, the World Meteorological Organization (MWO) have established the instruments technical characteristics and measurements protocols recommended for their determination (World Meteorological Organization, 2008). We followed those recommended practices. Measurements were reported as 1 hr-averaged values.

There is no real mention (Fig) about their measured data and its variability. Also, based on their 24-hr filter sample, how did they tell anything about traffic influence since traffic is very dynamic? With their 24 hour filter sample, they essentially do not have any temporal information. For example, the influence of meteorology (boundary layer variation), traffic (diurnal traffic variation).

Attending reviewer suggestion, we included in the manuscript figures describing the temporal variations of the measured data.

We did not oriented this manuscript towards the description of PM10 and TSP dispersion based only on our experimental measurements because it will limit the validity of our conclusions to the specific conditions and timeframes under which we developed our experimental work. We neither intended to correlate TSP and PM10 concentrations with traffic and meteorological conditions because we chose to study the influence of those variables on pollutant concentration through the physics of dispersion included in the NR-CFD model.

Again, the purpose of the experimental work was to validate our NR-CFD model. Then, we used the calibrated NR-CFD model to study the effect of the varying conditions of traffic on near road pollutant concentration, in terms of short and long term concentrations, so that they can be used to asses human health impact. Using 1 hour values for traffic conditions and meteorological variables, we modeled the dispersion of fine particles every hour assuming that steady state conditions prevail within each 1-hour time interval. Then we averaged those results for periods of 24 hrs and compared them with experimental measurements. We also compared modeled and experimental results in terms of 1 month averaged values. As stated in the manuscript we found high correlations among them

indicating the NR-CFD model and our approach of 1 hr modelling predicts well short (24 hrs) and long term (>1 month) concentrations.

Specific Comment 3. PM size distribution and composition: It is very confusing that they frequently generalized PM without mentioning any size information. What they measured is road dust (PM10 and TSP). Traffic emitted particles are dominated by smaller particles (a majority of combustion particle). What traffic-related info they might get based on filter SEM analysis of coarse PM? They reported that changes in particle size distribution are negligible within _1 km from the road edge, which is very confusing and miss-leading.
First, their measurements are mostly road-dust, not traffic particles, so there should not be any significant gradient for that. In reality, the size distribution of traffic-emitted particle in a near-road environment is highly dynamic and changes very rapidly within a few hundred meters from the roadway (Zhang and Wexler, 2004). Several complex microphysical processes dictate that changes, such as dilution, evaporation, condensation, coagulation, etc. Since they only measured TSP, which is not that traffic-related. Therefore, their results would not tell the true nature of the typical traffic-related particle.

Reply: *ok, manuscript was modified.*

It is very confusing that they frequently generalized PM without mentioning any size information
We use the following terminology regarding particles.

- *PM (Particulate matter): solid phase particles, regardless of their size, particle size distribution, morphology or chemical composition*
- *TSP (Total suspended particles or fine particles). Particles with aerodynamic diameter <30 μm,*
- *PM10: particles with aerodynamic diameter <10 μm,*
- *PM2.5: particles with aerodynamic diameter <2.5 μm,*
- *UFP: particles with diameters in the range of 1-100 nm*

What they measured is road dust (PM10 and TSP).
Yes. We also measured PM2.5. (Pg 5, line 5). As described previously, they are traffic related pollutants.

Traffic emitted particles are dominated by smaller particles (a majority of combustion particle).
In the general case, particles emitted from roads include: i.) Particles emitted from the vehicle tailpipe (exhaust emissions) ii.) Particle emitted due to wear and tear of vehicle parts such as brake, tyre and clutch iii.) and re-suspension of particles (non-exhaust emissions) (*Pant and Harrison, 2013*). *It has been shown that even with zero tail pipe emissions, traffic will continue to contribute to fine and ultrafine particles through non-exhaust emissions* (Briefs & Environmental, n.d.)(*Dahl et al., 2006; Kumar et al., 2013*) *and it is estimated that nearly 90% of the total emissions from road traffic will come from non-exhaust sources by the end of the decade* (*Rexeis and Hausberger, 2009*). *Non-exhaust emissions, are becoming more important now, and further research is anticipated in this field in the coming years* (*Pant and Harrison, 2013*).

What traffic-related info they might get based on filter SEM analysis of coarse PM
We did not intent to determine UFP concentration during our experimental work. As stated before, traffic related emissions include both: tailpipe emission and re-suspended particles.
SEM analysis confirmed that particle trapped in the filters came from the resuspension of particles of the same chemical composition that the road material. Based con SEM analysis we also obtained particle size distribution. SEM analysis did not show the presence of particles originated from combustion processes, which was expected because particle emissions due to resuspension is 4 orders of magnitude higher than tailpipe particle emissions.

They reported that changes in particle size distribution are negligible within _1 km from the road edge, which is very confusing and miss-leading. In reality, the size distribution of traffic-emitted particle in a near-road environment is highly dynamic and changes very rapidly within a few hundred meters from the roadway (Zhang and Wexler, 2004). Several complex microphysical processes dictate that changes, such as dilution, evaporation, condensation, coagulation, etc. Since they only measured TSP, which is not that traffic-related. Therefore, their results would not tell the true nature of the typical traffic-related particle.
Experimentally and trough simulation we obtained that particle size distribution for the case of fine particles ($1<d<30\ \mu m$) remains essentially constant within the first km from the road. As stated on the manuscript (pg 9, line 10), several other author have reached the same conclusion (Zhu et al., 2011). We modified the manuscript and clarified that this conclusion may not be true for the case of UFP.

We added a new paragraph discussing the dispersion of UFP and the results obtained by our NR-CFD model. See the end of this document.

Specific Comment 4. Traffic data (Page 4): how did they measure traffic data? Details should be given about measurement technique, data averaging time and data quality. Also, it is important to have some information about fuel use scenario (diesel vs. gasoline use). The reported traffic flow rate (20-50 veh./hr) looks very unreasonable to me, specially for an arterial road.

Reply: *ok, manuscript was modified. We included the additional information that reviewer suggested about the measurement campaign and data treatment related to traffic data.*

Yes, our traffic flow is too low for an arterial road. We did not state that we performed our experimental work near an arterial road. We did it near a local road and specifically near two unpaved roads (pg 3, lines 30). The purpose of our experimental work was to validate the NR_CFD model. We selected unpaved roads for this purpose because the procedures and instrumentation used to measure PM2.5, PM10 and TSP concentration are well stablished. Besides that, near unpaved roads PM2.5, PM10 and TSP concentrations are much higher that the uncertainties involved in the measurement procedures.

We also modified the title of our manuscript.

Specific Comment 5. P4: There are a bunch of equations, but there is no description of what are they and what is the meaning of different symbols. There is a list of symbols at the end, but it's good to have the description of symbol along with equation. Also, how did they get inputs for estimating EF, which is not clear to me? Clarification is needed.

Reply: *ok, the manuscript was modified. We included a description of symbols along with the equations. We also clarified on the data used to estimate EF.*

The following equations estimate the mass of fine particles emitted from paved and unpaved roads,

$$E = \frac{\sum N_j E_{fij}}{3600\ L}(1-\eta_r)(1-\eta_{rn}) \qquad (1)$$

where

E	TSP mass emission rate per road area	*g /s m²*
E_{fi}	Emission factor for vehicle of size i in kg of TSP per vehicle and per km traveled	*kg/VKT*
L	Road width	*m*
η_r	Efficiency of particulate matter emission control by rain	-
η_s	Efficiency of particle emission control by water spraying	-

The USEPA recommends to use the following E_{fi} for the case of TSP and PM10 emissions from paved and unpaved roads.

Table 1.
Emission factors for TSP and PM10 emissions from paved and unpaved roads.

Source		***TSP***	***PM₁₀***
Unpaved roads (US EPA, 2006)		$E_{fj} = 1381.31\left[\left(\frac{s}{12}\right)^{0.7}\left(\frac{W}{3}\right)^{0.45}\right]$	$E_{fj} = 422.85\left[\left(\frac{s}{12}\right)^{0.9}\left(\frac{W}{3}\right)^{0.45}\right]$
Paved roads (US EPA, 2011)		$E_{fj} = 3.23\left[(s_L)^{0.91}(W_j)^{1.02}\right]$	$E_{fj} = 0.62\left[(s_L)^{0.91}(W_j)^{1.02}\right]$

Where:

m	Number of rainy days in the period with precipitation levels exceeding 0.254 mm	*days*
n_d	Number of days in the period	*days*
N_i	Number of vehicle of size *i*	-
p	Average daytime evaporation rate	*mm/h*
q	Irrigation application intensity	*L/m²*
r	Average daily traffic	*Veh/h*
s, s_I	Silt content of road surface material	-
t	Average time between spray applications	*h*
W, W_j	Average weight of the vehicles of size j traveling in the road	*Tons*

The following equation estimate vehicle tail pipe emissions (Et).

$$E_t = \sum E_{fj}\ N_j\ T_j$$

Where

T_j	*Kilometers traveled by vehicle j*	*Km*

Exhaust pipe emissions factor (Eij) for tailpipe emissions are available in several databases (e.g. CEPMIEP 2014; NAEI 2014).

These parameters were measured hourly during the monitoring campaign. To do so, every time a vehicle crossed by, its average speed and type of vehicle was recorded. The type of vehicle was later associated to its approximated vehicular weight.

(Pg, 4, line 4) We determined road surface silt content using method INV E15 123 from the Colombian National Road Institute (Instituto Nacional de Vías, 2007). Silt content is the mass fraction of particles with diameters smaller than 200 μm present in the road surface material. We found that both roads had a silt content of 9.5%.

During the monitoring campaign there was no rainfalls ($\eta_r=0$) nor water was sprayed to control particle emissions ($\eta_s=0$)

As stated before, we found that fine particles emitted due to resuspension is by far the largest source of fine particles from unpaved roads (at least 4 orders of magnitude higher than tailpipe emissions).

Specific Comment 6. They reported that the non-dimensional concentration of all gas phase pollutants exhibits a unique profile (Figure 9.a) that can be represented by a beta function with parameters. This is something over-weighted (more generalized) to me. Can the author model the concentration profile from different seasons using their unique function? I'd expect a substantial seasonality on near-road pollutant gradients. Can their unique function account the seasonality and different physicochemical transformation of different pollutants as well? Did they test it? Otherwise, this conclusion might be very misleading.

Reply:

These are the major assumptions of our NR-CFD model:

- *At every hour, dispersion occurs under pseudo steady state conditions.*
- *Dispersion happens on a flat terrain with no obstacles to the dispersion of pollutants and the road is the only source of pollutants*
- *Temperature remains constant within the computational domain (~ 50 m of height)*
- *No chemical reactions (or phase changes) occur.*

We confirm that under these circumstances, the dispersion of all gas phase pollutants result in a unique profile of concentration vs distance to the road edge when expressed in terms of the non-dimensional numbers described in the manuscript. We also confirm that this profile can be described by a beta function with the parameters described in the manuscript.

We did test this observation under very diverse conditions of gas dispersion such as wind speed, emission rates and gas properties. We spent lot of effort looking for the set of appropriate variables that make the non-dimensional concentration profile unique.

Our results do account for seasonality. Even though the non-dimensional concentration vs distance profile remain the same, the actual (dimensional) gas concentration vs. distance changes with seasons. The main effect is due to temperature changes. Temperature changes diffusivity, density and viscosity of pollutants and gas-phase media. Even though atmospheric conditions change, the physics of dispersion remain the same.

Specific Comment 7. Vertical profile of PM distribution: Did they measure it? Can they evaluate their model results? TSP concentrations in an unpaved road would be highest at ground level that makes sense. But, for traffic emitted pollutants (e.g., ultrafine particles), it could be very different. They should be very careful while reporting different PM fraction. They should not generalize PM without any size information. This is very confusing throughout the paper.

Reply: *Ok, we modified the manuscript to clarify the type of particles that we are referring to.*
We did not measure any vertical particle concentration profile. We compared qualitatively our results on TSP vertical concentration profile with experimental results reported in the literature (Yuan, Ng, and Norford, 2014;Shen, Cui, and Zhang, 2017; Kwak, Baik, Ryu, and Lee, 2015). We found that they exhibit the same profile.

See the end of this document for results on the vertical profile of UFP obtained by our NR-CFD model.

Specific Comment 8. P10, L47: "we used the NR-CFD model to study differences in the dispersion of CO, CO2, NO2 and TSP"- Did they measure these gases? There is no description on that?

Reply: *Ok*
We did not measured these gases. In P11, L7, we stated "Aiming to validate these results, we looked in the literature for experimental data. Several works have reported measurements of gas phase pollutants concentration near roads. However, none of them reports simultaneous measurements of mass emissions, meteorological conditions and pollutants concentration that could be used to validate quantitatively the NR-CFD model. As an approximation, we compared, qualitatively, NR-CFD results to values of NO2 measured near roads with high traffic of heavy-duty vehicles in UK (Chaney et al., 2011). Each set of simultaneous NO2 measurements were normalized in the way that the area under the concentration vs distance to the road edge curve were equals to one. Figure 9.b shows that numerical and experimental results are similar. Non-dimensional concentration differs from normalized concentration in their area under the curve but their shapes are similar.

Specific Comment 9. P5L1: "primary and secondary meteorological variables"- not sure what did they mean by primary and secondary met variable here? Which are primary and which are secondary?

Reply*: Ok. Manuscript was modified. We removed "and secondary" from the manuscript.*

Specific Comment 10. "Particles exhibit a Rosin Rambler size distribution with average diameter of _ 7 _m" – This is again very confusing. What did they mean by particles here? Particle mass or number size distribution? It seems PM mass. However, how relevant is this in context of traffic-emitted particles? I guess, this is only telling something about road dust, not much about traffic-emitted PM. Clarification is needed.

Reply:
Manuscript was modified.
We referred to particle number size distribution. We obtained it counting particles observed in SEM photographs.
References

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Best Regards
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