The authors would like to thank the reviewer for their careful review. We have attached answers to their specific concerns here and have also uploaded a merged document showing tracked changes we have make within the manuscript in response to their concerns.

This paper presents analysis of the data from an approximately 2 weeklong measurement campaign at a site adjacent to a large freeway outside Grenoble, France. An extensive set of real-time instrumentation for measurement of particle- and gas-phase species was used including an SMPS for particle size/count, an HR-TOF-AMS for particle composition/size, a MAAP for BC light absorption, a TEOM for PM mass (though I did not see mention of these data), ***(PM data is included in the supplementary information and mentioned in the text)*** a PTR-TOF-MS for VOC measurement and a NOx analyzer. Integrated samples were collected for multiple analyses, including carbon isotope analysis to determine the modern/fossil fraction of total particulate carbon (TC). Supporting measurements included detailed traffic flow, speed and composition measurements and a meteorological station to measure wind parameters, temperature and RH. The analysis presented in the paper is similarly comprehensive and includes: examination of diurnal trends, linkages with traffic conditions, PMF decomposition of AMS data and factors' relations with other measurements (e.g. VOCs, NOx, BC), and analysis of the fossil/modern carbon fraction of the total carbon.

This is a large and comprehensive dataset, and it appears that the data collection and analysis were well planned and executed. These data have the potential to provide valuable insights into the characteristics of emissions from a large and diverse vehicle fleet and to their contribution to urban/regional air pollution. Further, the title of the manuscript indicates that this is the goal of analysis. My major issue with this analysis is that it does not proceed in a way that can truly address this objective. Rather, as presented, the analysis treats the data set as a 'standard' ambient monitoring study, that pays little attention to the relationship between the source in question (the roadway) and transport of these emissions to the point of measurement (e.g. wind direction). If this work were presented as a routine monitoring effort at a site that happens to be near a large road, I might not take issue with this presentation. However, given that 'emission' is in the title, I expect to see more of a focus on trying to find the 'emission' signature, rather than just expecting it to be there and constant during all measurements. Therefore if the title were something like 'Comprehensive gas- and aerosol-measurements collected near a roadway in a high-diesel peri-urban environment',
I would expect this kind of analysis. However, characterization of emissions should make efforts to separate or characterized the background vs roadway contributions.
So, one approach could be to change the title of the paper and apply some of the changes suggested here and by the other reviewer. However, to actually address the objective implied by the title, more work needs to be done to extract the roadway 'emission' signature from the data. My sense is that not doing this additional analysis would be a missed opportunity.

We understand the reviewer's point and agree that we have not provided enough emission calculations to have 'emission' in the title. We have both changed the title of the paper to reflect the reviewer's concerns and also included more information on traffic-heavy periods within the paper (particularly in Figure 2 and Figure 7, discussed further below).

There is a rather extensive literature of near-road air pollution measurements ((Karner et al. 2010) is a good place to start), which seems to be largely ignored in this analysis aside from the few studies in which an AMS was used. The important message from
these previous studies are that concentration gradients are: a) steep near roadways, b) a strong function of wind direction/speed and mixing conditions. However, the spatial and meteorological aspects of this data set are entirely absent from the analysis as presented.

Unfortunately, the measurement station was fixed in location, so gradient changes were impossible to determine, but we have now added more details in Figure 1 and discussed in more detail the wind direction, wind speeds, and location of the highway relative to the measurement site. Supplementary information included polar plots showing mass loadings of different AMS factors as a function of wind direction. As noted at the beginning of section 3.1, wind speed was low and the station location was noted both in words and in Figure 1.

Now, following the reviewer's suggestion, Figure 1 has been updated to include more details on the measurement location and now includes polar plots of BC and NOx as well as a wind rose to make the meteorological conditions more clear. Section 3.1 includes this description of the meteorological conditions at the sampling site: Wind speeds were generally low throughout the campaign (<1-2 m/s) with higher wind speeds peaking in the afternoons and tapering off in the evenings. The wind direction was primarily from the northwest, from the direction of the nearby highway. A diagram of the two measurement stations, the wind rose plot for the Massalya location, and polar plots showing the concentration of NO and BC as a function of wind direction are shown in Figure 1, along with a map of the local area and the position of Grenoble within France. " There is also already a discussion of boundary layer conditions, which has a large effect on aerosol concentrations.

We have now cited the suggested paper and included this sentence about gradients "The fixed location of the measurement stations made determination of concentration drop-off as a function of distance from the roadway impossible to determine with the dataset, although that has been shown to be important in other studies (such as Karner et al., 2010). However, the measurements were all taken closer to the roadway (~15 m) than the calculated distance where roadway emission drop off to background levels (115-570 m, Karner et al., 2010)."

The air sampled at this site will be a combination of the background/regional mixture (heavily influenced by other roadways with similar diurnal various to traffic and fleet among the multitude of other sources) and emissions from vehicles on the adjacent roadway. Strength of having all of these highly sensitive, high-time-

resolution instruments is that the authors should be able to pick the 'emission' signal out from the background, by sorting data based on wind speed/direction. It could be that due to the low wind speeds observed in this study (I cannot comment on wind direction, as no data were shown, nor was the relationship of the measurement site to the roadway)
that is a small portion of the overall data set, but it would still get at the 'emissions' question.

We thank the reviewer for pointing out the need for more detail and clarification regarding the meteorology of the sampling site. This was discussed in the section 3.2 General Atmospheric Conditions: "Wind speeds were generally low throughout the campaign (<1-2 m/s) with higher wind speeds peaking in the afternoons and tapering off in the evenings. The wind direction was primarily from the northwest, from the direction of the nearby highway." As per the reviewer's suggestion, we have added a wind rose, polar plots as a function of wind direction of BC and NO in addition to the polar plots of AMS factors previously included in the supplementary information, and a more detailed diagram of the measurement site to hopefully make the meteorological conditions more clear.

Starting on Line 313 in the merged/revised document with tracked changes, we now include this paragraph (some repeated from the past version): Wind speeds were generally low throughout the campaign (<1-2 m/s) with higher wind speeds peaking in the afternoons and tapering off in the evenings. The wind direction was primarily from the northwest, from the direction of the nearby highway. A diagram of the two measurement stations, the wind rose plot for the Massalya location, and polar plots showing the concentration of NO and BC as a function of wind direction are shown in Figure 1. BC and NO were associated with all wind directions, though slightly higher from the highway direction, which suggested that the measurement site was directly and strongly influenced by the traffic emissions. However, in order to better describe the traffic influence, we defined high traffic periods (HT). These HT periods were selected as follows : wind direction >40 or <320, NO in the 75th percentile, and from 6:30-9:30 or 17:00-20:00 (rush hour periods). The fixed location of the measurement stations made determination of concentration drop-off as a function of distance from the roadway impossible to determine with the dataset, although that has been shown to be important in other studies (such as Karner et al., 2010). However, the measurements were all taken closer to the roadway (~15 m) than the calculated distance where roadway emission drops off to background levels (115-570 m, Karner et al., 2010).

Based on the factor-based wind roses in Figure S8, I would hazard a guess that the roadway was to the North/Northwest of the sampling location give the asymmetry in the HOA contribution on the wind rose (correction – just found the mention of this in Sec. 3.2). I would expect that the authors could extract portions of the data set that allow them to more directly evaluate traffic emissions, ideally by contrasting with other periods where the measurements were upwind from the roadway. Other measurements

from regional air quality monitoring stations might be of use in assessing changes in regional levels to extract the roadway signal. The existing analysis can be presented along a more directional approach; there are likely valuable insights gained from each approach.
We have now defined a period of time as high traffic, based on wind direction, NO concentrations, and times correlating to morning and evening rush hour. These HT periods were selected as follows : wind direction >40 or <320, NO in the 75th percentile, and from 6:30-9:30 or 17:00-20:00 (rush hour periods). More detail is provided both in Figure 2 and Figure 7 (comparison of the relative contribution of AMS organic/inorganic species and BC, and PMF factors, respectively, for all and high-traffic time periods. PM2.5 and PM10 increased slightly during HT periods (a 1.25 and 1.3x increase, respectively).BC and m/z 57 increased 1.5x, benzene by 20%, CH4NO2 by 8%, and 1.9x increase for HOA and 1.3x increase for NOA PMF factors.

Apart from this framing/objectives concern, the main other concern I have is concerning the analysis of the fossil/modern carbon fractions. This analysis needs more attention, and I find it somewhat difficult to follow the process described, the results shown in Fig. 15 and the conclusions of this analysis. For one, a number of assumptions appear to be made in the process of apportioning the modern/fossil contribution to OA, but these are not at all clear. It appears that the modern C contribution to EC/BC is ignored, as is the potential modern C contribution to HOA, but I may not be clear on this. Given the statement that as much as 7% of diesel may be biodiesel and that BBOA made a reasonably large contribution (Fig. 10b) to the OA loading at times, these assumptions seems to be potentially biasing and also unnecessary (given all of the other supporting data/analysis that is presented). I would like to see this process better spelled out and the uncertainties in the various assumptions required apportion the modern/fossil C better constrained and the impact of measurement uncertainties and uncertainties due to assumptions propagated through to this overall apportionment exercise. In particular, the measurement of EC/BC is a potentially large source of uncertainty that would introduce an enormous bias into this analysis. That the light-absorbing carbon is discussed as BC suggests that these quantities are based on an optical measurement, and there is huge variability/uncertainty in the absorption cross-section and thus the BC concentration measurement generated by something like a MAAP. There is mention of a thermo-optical analysis of filters, I think mainly from the other site, but no data are ever shown. Ideally, all of these comparisons would be on a mass basis (i.e. EC, not BC) and the uncertainties incorporated into the analysis. If these various uncertainties are addressed, a more quantitative apportionment of C mass will be possible, giving the discussion of SOA sources/etc. a more solid footing. In addition, Fig. 15 is not clearly labeled – preferably segments would be labeled as either OC or EC to avoid any ambiguity.

We thank the reviewer for their corrections. We used EC data for the calculation of this figure and have now correctly labeled it as such, and thank the reviewer

for pointing out this error. Additionally, we compare EC (Sunset Laboratories thermal analyzer) and data from the MAAP instrument (optical) in the supplementary information. These instruments were not located at the same station (EC at the Air Rhone Alps station, MAAP within the Massalya platform) but agree fairly well and show no large higher bias in the MAAP data until higher loadings. Additionally, a range of fossil-OOA has been calculated. We have also re-labeled the graph, hopefully making the values presented on the graph clearer.

We have added significantly more detail about the calculation in section 3.4.1, including sources of error and discussion of possible biases in data. As the reviewer suggested, we include a range for possible modern-C from the EC and HOA factors to account for the potential contribution of biomass burning and biofuel use to these species. While biofuel is thought to emit less particles/car than traditional diesel vehicles, we agree it is important to include the possibility of biofuel contribution to EC and HOA in this calculation.

Following is the revised section, beginning on line 731 in the merged document:

> *A source of uncertainty in the global particulate emissions of vehicles is the formation of SOA from gas-phase emissions and the aging of POA. To discriminate between the relative concentration of modern and fossil carbon, and thus potentially discriminate between OOA from vehicular sources and from modern sources, daily filter samples were collected at the sampling site and 14C radiocarbon measurements were performed. From these measurements, the percentage of modern carbon from TC (OC+EC) was calculated. Modern carbon varied from 15-36% of the total aerosol carbon, a significant portion of the measured carbon considering the close proximity of the measurements to fossil carbon sources. In France, the contribution of biofuel was about 7% and 5% for diesel and gasoline fuel, respectively, in 2011 (UFIP, Union Française des Industries Petrolières, 2011) and cannot explain this relative high proportion of modern carbon observed in the particulate matter. This is similar to findings shown in Hodzic et al. (2010), Minguillon et al. (2011), and El Haddad et al. (2013), which indicate that modern carbon is often more significant than fossil carbon in the carbonaceous fraction of PM, even in cities with high vehicular emissions (e.g., Mexico City, Barcelona or Marseille).*
>
> *As radiocarbon measurements have been performed through a thermal approach (combustion of the samples at 850°C), we consider in the following section EC measured by the thermo-optical method. As shown in figure S12, EC and BC do not differ significantly at low mass loadings, but have a wider scatter in the data at higher mass loadings. The calculation of BC (measured by the MAAP) using an absorption cross-section is imprecise and, at high loadings of BC, may under or overestimate this mass loading. Figure S12 shows a comparison between the MAAP (BC) and thermal measurement (EC) data, with a 1:1 line. As the thermal-optical analysis of EC is a more direct analysis, EC was chosen to be used in this calculation.*

Assuming that the majority of EC was traffic-related, and thus from fossil origin, the concentration of modern organic carbon and fossil organic carbon was then calculated. While evidence for the presence of biomass burning aerosol was measured at the field site, the main source of EC was likely diesel exhaust. Figure 8 shows the fraction of EC and OC, HOA, and a partitioning between fossil and modern carbon. In Figure 8A, a rough calculation was performed to determine the concentration of non-primary fossil organic carbon. For a first estimate, all EC was assumed to be fossil in origin. Additionally, the HOA aerosol was also assumed to be vehicular, and thus fossil, in origin. The HOA factor concentration has been divided by its OM: OC ratio to remove any non-carbon mass (HOA C, calculated from the elemental formulas of the PMF factor mass spectra, Aiken et al. (2008)). Both EC and HOA C had high (R^2=0.89 and 0.85 respectively, n=10) correlations with the fossil C mass, which supported a largely fossil source for these two species. The remaining fossil organic carbon mass was then assumed to be from non-primary sources.

This calculation provided a lower estimate of the amount of fossil carbon contributing to SOA mass, and involves several assumptions and potential sources of error. Sources of error in this calculation include error in the PMF resolution of primary (HOA) organic aerosol spectra and error in the calculated OM:OC ratio of this factor species, biodiesel vehicular emissions contributing modern carbon to measured HOA, and biomass burning aerosol contributing modern carbon to measured EC.

As the measured HOA:EC ratio was in-line with previous measurements in high diesel environments, HOA concentrations did not appear to be significantly over or under estimated. Up to 7% of fuel use in France was biodiesel, thus, part of the HOA concentration could be from modern sources. While research has shown that the use of biodiesel fuels reduces the overall primary particulate matter emissions (Cheung et al, 2010), biodiesel could still be a modern carbon contributor to OC and EC mass. Additionally, although the concentration of BBOA was generally low (a campaign average of 0.34 ± 0.23 μg m^{-3}) and the ratio of BBOA:EC has been found to be on the order of 3-4 in other areas of France (Crippa et al., 2013), some contribution to EC from biomass burning may have been present at the measurement site. In Figure 8B, a range of fossil non-primary organic carbon, normalized to total measured organic carbon, is presented. For the upper limit of this range, HOA C and EC were considered to be 95% fossil and 5% modern (7% biodiesel fuel use and an estimated 25% reduction in particulate emissions from biodiesel fuel). Also for this upper limit, the calculated concentration of BBOA was divided by 3 and used to calculate possible modern EC from biomass burning (Crippa et al., 2013).

Total organic carbon concentration appeared to be more driven by processed/aged OOA concentrations than by primary emissions. During the period with the highest organic concentrations (September 15th-17th), most of the non-HOA carbon measured was modern carbon. Also during

> ***this time period, the winds were also slightly more southerly and SO_4 and OOA concentrations increased, which could indicate a more regional contribution to the measured air mass during this time. After a period of heavy rain on the 19th, almost none of the non-HOA, organic carbon was fossil; however, this also coincided with a period of increased BBOA, which may have contributed to modern-EC emissions and thus an underestimate of fossil-OC emissions (Figure 7). At other times during the campaign, HOA concentrations alone could not adequately explain all of the measured fossil organic carbon and additional sources of fossil organic carbon (such as photochemical reactions forming aerosol from vehicular VOC emissions) would be needed. Additionally, the origin of the NOA factor remains unclear, and if fossil in origin, could explain part of the non-HOA organic fossil carbon measured at the site, further reducing the OOA fossil-C (at times to almost zero).. Overall, throughout the campaign the majority of OOA observed was most probably modern in origin.***
>
> ***The high levels of modern carbon OOA suggested that biogenic compounds had the greatest effect on the overall aerosol population in this location, even directly adjacent to a large anthropogenic emission source (i.e., traffic). Interaction between anthropogenic oxidants and biogenic VOCs (or BVOCs) has been found to increase the formation of SOA (Chameides et al., 1988; Goldstein et al., 2009; Shilling et al., 2013), isoprene oxidation reactions leading towards SOA have been shown to vary depending on the level of NOx (Chen et al., 2014; Kroll et al., 2005; Ng et al., 2007; Xu et al., 2014), and likely BVOC concentrations were greater and the aromatic VOC concentrations were lower in the wider Grenoble Valley.***

Finally, while I am suggesting additional analysis, I agree with the other reviewer that the number of figures is excessive. Figures 2, 6, 11, 12 and 14 could easily be moved to SI without diminishing the readability of the manuscript.

These figures and the discussion of these figures were moved to SI as suggested, there are now 9 figures in the manuscript.

Other specific points: P27376, L15-18 – Somewhat nitpicky, but the use of 'rate' here is not really appropriate, as this really depends on engine size. You could say 'emission factor' or 'ratios'. . .

Changed to factor, as suggested.

P27376, L25-30 - Suggest reorganizing this paragraph as there is a transition to discussion of engine-type-specific emissions to results of several specific field measurements.

We have removed some of this paragraph, which hopefully will make the transition smoother.

L6 – There has been no previous mention of smog chamber studies, so this way of starting the paragraph is a bit confusing.

We have removed this line.

P27377, L16-30 – Not so clear how this particular paragraph pertains to the study objectives. Seems like it could be replaced with a sentence stating that there is controversy concerning the relative contributions of gasoline and diesel engine emissions to primary and secondary OA in the US.

This relates somewhat to the fossil v modern carbon OOA calculations later in the paper, but this paragraph has been shortened as per the reviewer's suggestion.

P27378, L17-20 – This seems like a major objective of the study, but then there is very little comparison (apart from e.g. HOA/BC ratios. This could be highlighted more and then more fully addressed.

The reviewer is right, this was one of the goals of the study. Also compared are NOx measurements, HOA chemical composition (unfortunately the same for diesel and gasoline), and partially relates to the fossil or modern-carbon OOA (as previous studies have suggested that VOC emissions from gasoline vehicles can lead to more SOA formation than those from diesel).

To help draw this point more clearly, the authors have moved the discussion of these three things into a new section "3.4.3. Differences between diesel-heavy and gasoline-heavy near-roadway measurements" where we discuss in more detail the comparison of gasoline-rich and diesel-rich near-highway measurement campaigns.

P27379, L19 – This seems like a natural place to discuss the meteorology of the site, location of measurements relative to roadway, prevailing wind directions, etc.

We now include a more detailed figure (Figure 1) including a wind rose.

P27382, L13 – It is not clear where this site is relative to the mobile lab, how these are impacted differentially by wind, and where data from the two sources are used in concert in this analysis. Were there any duplicate instruments on the two platforms on which this comparison can be made? In general, the contribution of data from the different sites could be better delineated.

The aerosol and VOC measurements were from the MASSALYA mobile lab, while the gas-phase NOx measurements and filter samples were taken at the Air Rhone Alps station, as detailed in their sections. No exact overlapping measurements were collected; however, the EC measurements from the Air Rhone Alps station are now compared with the BC MAAP measurements from the Massalya platform in the supplementary information as an answer to another question, and agree fairly well. Both measurement sites were close and on the same side of the highway, as is now more clearly shown in Figure 1. They were 20 m apart (Air Rhone Alpes station 20 m to the east).

P27383, L5-18 – This discussion of the fleet composition on this roadway is only really directly relevant if you are measuring emissions from these vehicles (see discussion above). Otherwise, you are better served by talking about the regional fleet composition (and other potential sources). This is a very detailed description of the source term, but is not relevant if you're not sampling from this source.

We have now moved the fleet composition graph into the supplementary information and have removed more of the detailed discussion of the fleet as per the reviewer's suggestion.

P27384, L5 – The only mention of meteorology. There should be data accompanying this, including a more detailed diagram of the placement of the road, mobile lab and the regulatory monitoring site.

There is now more detail regarding meteorology (a wind rose) and mobile lab placement in Figure 1, as well as a statement on the wind speeds. The proximity of the highway and the low wind speeds, as well as close proximity of other highways, made the meteorology less important than it would have been in perhaps other measurement locations with higher wind speeds and/or more variable wind speeds and directions.

P27384, L21 – BC measurements need to be defined (e.g. comparison with EC, etc.)

Now there is a comparison between BC and EC in the SI.

P27385, L6-7 – The Org/BC ratio is much lower during this period as well. There seems to be an opportunity to analyze specific periods/episodes to try to better extract the vehicle signature from other things. The BC variation is relatively consistent, but the 15th-18th has a much larger OA fraction and more of these other factors, and also much lower NO, this seems to be a different type of period than the rest of the study. Can you use differences between periods to infer the difference between roadway/regional contributors to measured levels?

The reviewer is correct that, during this time period, more regional aerosol was measured at the station site. That was one way we were able to separate out regional OOA (in the middle period, with high Org, lower BC/Org ratio) and discuss regional contributions. We now have added the statement "The slow rise of organic concentration during these periods, the lower BC/Org ratio, the enhancement of organic concentration outside of normal traffic periods, and the low level of NO during these accumulation periods all suggest that this increase in organic aerosol concentration was driven by regional influences, not by nearby vehicular emissions." (line 426 in merged revised document).

P27386, L6 – this is not really 'BLH-corrected', as it's not clear that's something you can correct for. In any case, the rationale and approach could be better described. What is the motivation for this approach versus just plotting the diurnal variation in ratios(e.g. benzene: BC or some other primary vehicle emission tracer).

Yes, we agree with the reviewer that this terminology should be changed. We have now changed the wording to BLH-scaled.

The motivation behind scaling for boundary layer height was to more directly compare vehicle concentrations with aerosol/VOC concentrations on an hourly basis. We chose not to scale to benzene or BC as VOC and aerosol emissions change both with vehicular concentration and with vehicle running conditions (idling, start/stop, typical highway driving). By performing this rough scaling calculation using an estimated BLH, differences in the concentrations of vehicular-related species as a function of both vehicle numbers and vehicle speeds was more apparent (and thus VOC and aerosol response to traffic speed and concentration could be more readily compared).

P27387, L27-28 – The evidence for this is not really shown, also need to show the mass in each size bin and the contribution of HOA/OOA to each size bin. Also, I'm not clear of showing the average values for the entire campaign. This represents a snapshot of the climatology of the local aerosol, but this analysis might be more useful if it compares times with different roadway vs. non-roadway contributions.

Right now it would be very time consuming and potentially fraught with error for the authors to separate HOA and OOA in each size bin using their PMF model. M/Z 44 and 57 are commonly used as markers for OOA and HOA, respectively, in AMS data. The authors have moved this graph and the accompanying discussion to the supplementary information, as it largely agrees with previous findings that primary organic vehicular aerosol (apart from resuspended road dust) is smaller than aged/secondary aerosol (e.g., Sun et al., 2010).

P27998, L26 – 'was SOA' is a very definite statement. . .

Changed to was OOA.

Figure S2 – It is hard to see in this presentation, but it appears that the PM data (presumably from TEOM?) has substantially more variation than the AMS data. Was the mass estimate from the MAAP benchmarked against the EC data? A scatter plot would be helpful. What about comparisons with SMPS volume data.

A scatter plot of MAAP and EC data is now shown in figure S13. The PM data is from the TEOM, yes. Likely the different size cutoffs (1 micron for the AMS versus 2.5 microns for the TEOM) is a large cause of the variability, as well as the difference in what each instrument can measure (the AMS cannot measure refractory species). A few larger road dust particles, for example, could drive much of the mass-variability in the TEOM data that would not be seen in the AMS data.

The SMPS in turn both has a different size cut-off than the TEOM instrument and measures both refractory and non-refractory compounds, a difference between SMPS measurements and AMS measurements. Road salt, dust, etc. would be measured by both the TEOM instrument and the SMPS instrument, but not by the AMS or the MAAP. As the general time trends are the same for both the TEOM instrument and the AMS instrument, we believe they were capturing similar events but their absolute concentrations are not directly comparable.

We now explain this in the text:

Line 392 "PM2.5 has a somewhat higher mass variation than the AMS + BC measured mass (Figure S3), likely due to the smaller measurement size cutoff for AMS (1 μm) and the presence of road dust in the local environment, a large

portion of which may be non-refractory and those unable to be measured by the AMS. "

References: Karner, A. A., Eisinger, D. S., and Niemeier, D. A. (2010). "Near-Roadway Air Quality: Synthesizing the Findings from Real-World Data." Environmental Science
& Technology, 44, 5334–5344.