Response to Reviewer's Comment

(Manuscript No. acp-2014-359)

Anonymous Reviewer #1

General Comments:

In this manuscript, the authors present results from a field study conducted at the Hsuehshan tunnel in Taiwan. Size resolved particulate matter samples were collected near both the entrance and exit of the tunnel on twelve days in 2013, and subsequently analyzed for total PM mass and metals content through gravimetric and ICP-MS analyses, respectively. Metal size distributions, enrichment factors, element ratios and results from principal component analysis are presented and used to explore sources of metallic aerosol components in the tunnel including tailpipe and wear emissions and resuspended dust. The authors also provide estimates of emission factors for trace elements and PM. In general, the scope of work pursued here is good and the authors have collected a nice data set that contributes to our understanding of vehicle derived trace metal aerosols. With some exceptions (see specific comments below), the manuscript is relatively well written and figures and tables are adequate. This being said, there are some major issues with the manuscript related to emission factor quantification that need to be addressed prior to publication in ACP. Specific comments on the manuscript are included below.

Author's response:

We have followed the reviewer's comments and revised this manuscript. The revised portion will be explained in details of the following response.

1st comment

P. 13966, line 13-24: The authors provide a list of a number of possible sources of vehicle derived metal emissions. I wonder if this information would be clearer to the reader if presented in tabular format.

Author's response:

As suggested, we have added the new "Table S1" in supplementary to list the potential sources of these metallic elements in different sized PM.

2nd comment

P. 13967, lines 2-5: While I agree with the authors that tunnel studies, as compared to

near-road sampling, provide a more constrained environment for investigation of vehicle derived PM emissions, I think the authors need to do more here to substantiate the claim that near-road studies are "insufficient". For example, Ning et al. show that emission factors for several metals can be obtained from near-road sampling. (Ning, Z., A. Polidori, J. J. Schauer, and C. Sioutas (2008), Emission factors of PM species based on freeway measurements and comparison with tunnel and dynamometer studies, Atmos.Environ., 42, 3099–3114, doi:10.1016/j.atmosenv.2007.12.039.)

Author's response:

Thanks for the reviewer's comment. In the revised manuscript, we have cited additional references, such as Jamriska et al. (2004), Ntziachristos et al. (2007) and Ning et al. (2008), to claim the limitations of dynamometer tests and near-road side sampling in characterizing traffic-derived PM. The freeway measurement was done very well by Ning et al. (2008). Such airborne metals, including Cu, Mo, Ba and Pb, showed very good agreement with those measured by tunnel and dynamometer studies, but Mg, Fe and Ca did not. The disagreement was probably because the atmospheric concentrations of Mg, Fe and Ca are contributed by both traffic and non-traffic sources, and the relative abundances of these crustal elements in road dust varies considerably across locations. Moreover, some studies have corroborated that PM concentration near-road side measurements are influenced by local meteorological conditions and traffic activities (Jamriska et al., 2004; Ntziachristos et al., 2007). Thus, tunnel experiment may be an alternative way to characterize the traffic-derived PM. (lines 7-14 on page 4)

3rd comment

P. 13968, lines 16-24: The authors describe the ventilation system utilized in the Hsueushan tunnel here. It is unclear what the status of fans at exchange and inter-change stations was during the aerosol sampling campaign. The authors state that the fans are triggered when temperature or CO concentration thresholds are reached. Were the fans operational during the measurement periods? If so, the additional airflow into and out of the tunnel bore complicates the mass balance used to derive equation2 (see comment on Section 2.4 below).

Author's response:

Thanks for the reviewer's comment. In fact, more than 20 monitoring stations record the CO concentrations and temperature inside Huehshan Tunnel. The 1-hour average

CO concentrations at these stations never exceeded 50 ppm during the campaigns, indicating that the ventilation system was not triggered due to the high CO conditions. Nevertheless, the temperature inside the tunnel was usually above 40°C at the outlet site, especially, during July and August sampling period. This reflects that ventilation system was operated via high temperature. The operation of ventilation system resulted in the exchange between outside-air and inside-air inside Hseuhshan tunnel, leading to underestimated EmF of PM and their metallic elements of this work. Thus, we have deleted all the descriptions which are related to EmF in the revised manuscript.

4th commen

p. 13969, lines 2-3: I'm curious to know why the author's selected Friday-Sunday for their sampling periods. Many vehicle emission studies I am familiar with tend to prioritize weekday sampling, as traffic patterns on weekends and particularly on Sundays may not be representative. This study design is heavily weighted to weekend sampling. Some additional text here may be helpful. What are typical weekly traffic patterns at the tunnel? Is there any change in the overall traffic volume or vehicle mix on weekends relative to weekdays?

Author's response:

Thanks for the reviewer's comment. The traffic volumes inside Hsuehshan Tunnel are very different between workdays and weekends (particularly on Sunday evening). As listed in Table 1, the traffic flow at the northbound on Sunday was usually more than 1800 vehicles per hour, which was 1.3 times higher than that on Friday. The very high traffic volume (>2300 vehicle h^{-1}) was always found after 5 p.m. on Sunday afternnon, causing a traffic jam when lost of people traveled back to Taipei. The traffic jam does influence the characteristics of traffic-related PM metals. That is why we conducted the aerosol measurements inside Hsuehshan Tunnel from Friday to Sunday in each sampling campaign. (lines 16-24 on page 6)

5th comment

P. 13969, line 5: Please include location of sampling inlets within the tunnel bore crosssection (i.e. at what height were the inlets located).

Author's response:

As suggested, we have added the height of sampling intakes of both MOUDIs. (lines 14 and 15 on page 7)

6th comment

P. 13970, line 6: I could not find the method detection limits referred to in this sentence in Table S1.

Author's response:

We have added the method detection limit of each analyzed metal in Table S2 in the revised supplementary.

7th comment

Section 2.4: In regards to the calculation of emission factors using Eq. 2, I share many of the same reservations expressed in the short comment posted by Dr. Ceburnis. As presented, Eq.2 does not account for dilution caused by additional sources of airflow into and out of the tunnel between sampling points and likely results in an underestimate of emission factors. I see this as a major shortcoming of the manuscript and agree with Dr. Ceburnis that the authors need to better justify the appropriateness of Eq.2 as currently applied to evaluating emission factors in the Hsuehshan tunnel. Else, I suggest removing discussion of emission factors from the manuscript. Equation 2: Please include source of wind speed and traffic flow data.

Author's response:

Thanks for the reviewer's comment. We make sure that the ventilation system inside the tunnel was operated during the experiment campaigns, particularly in July and August experimental periods, resulting in air-exchange between polluted (inside the tunnel) and cleaner air (outside of the tunnel) and leading to underestimate significantly the EmF of PM mass and metals. Thus, we have removed all the descriptions related to EmF in the revised manuscript.

8th comment

Page 13972, lines 1-2: The argument that enhancement of carbonaceous material in submicron PM may be caused by absorption of organic gases by Teflon filters is speculative and unconvincing. Unless further support for this claim can be presented, I suggest removing.

Author's response:

As suggested, we have removed the sentence of "Another plausible explanation…..in submicron PM (Cabada et al., 2004)" in the revised manuscript.

9th comment

Page 13972, lines 4-21: In this paragraph, the authors discuss measured outlet to inlet mass concentration ratios of the three PM size fractions considered in this study. The authors assume minimal depositional losses of submicron PM and suggest the measured concentration ratio for this size fraction (4.4) should also apply to larger size fractions if no losses occur. Lower measured concentration ratios for PM1-1.8 (2.3) and PM1.8-10 (1.1) are then used to support quantification of particle losses in the tunnel for these larger PM size fractions. I find this line of reasoning problematic. It is not clear to me why a constant ratio for all size fractions should be assumed. A rearrangement of Eq.2 shows this ratio is dependent on the concentration of a given size fraction at the tunnel inlet and the emission rate within the tunnel: Co/Ci =(EmF×N×L)/(Vair×Ci)+1. Different processes dictate emissions of PM in the three size fractions and a constant emission rate should not be assumed. While the authors are correct that dry deposition will more efficiently remove larger particles, the treatment and quantification of these processes in this paragraph is flawed.

Author's response:

In this work, we used a factor of 4.4 as a reference ratio to correct the O/I ratios of $PM_{1\text{-}1.8}$ and $PM_{1.8\text{-}10}$. We agree that it is problematic since different process contributes different sized aerosols in real ambient conditions. Thus, we have re-organized this part in the revised manuscript. (lines 4-13 on 10)

10th comment

P. 13976, beginning line 19: I found the discussion of PCA results a bit confusing and difficult to follow. I suggest revision of this paragraph to more clearly explain interpretation of PCA results and relation between statistically determined principal components and potential sources. Some specific comments: -One main question I have is in regard to treatment of gasoline and diesel tailpipe emissions. In the fine PM group, PC1 is associated wear debris, dust, and gasoline emissions. PC2 is associated with "tailpipe emissions" which seem to refer to emissions from diesel engines based

on the text of this paragraph. First, I think the “tailpipe emission” potential source entries in Table 3 should be reworded to clarify whether the authors link the corresponding principal component to gasoline tailpipe emissions, diesel tailpipe emissions, or both. Also, the authors do not present enough evidence in the manuscript as to why different metals emission profiles should be expected for these two sources. Are different metal associated with these two fuels? Are lubrication oil formulations and additives different for gasoline and diesel engines? I think some additional text/references here would be helpful for the reader. -What was the fraction of diesel vehicles in the tunnel during sampling? In the site description section the authors explain that only passenger vehicles, light-duty trucks, and shuttle buses are allowed in the tunnel, with heavy-duty trucks (typically a major source of diesel emissions) presumably prohibited. A better documentation of the fraction of gasoline and diesel vehicles in the tunnel during sampling periods may improve the identification of potential sources for principal components. I am confused by PC4 in the submicron fraction which is attributed to “fuel oil”. Are the authors referring to diesel fuel here or are they referring to heavier fuel oils typically burned in larger diesel engines (e.g. marine engines). If the former, how is this a distinct source from the “tailpipe emission” component? If the latter, are the authors suggesting a contribution to PM in the tunnel from a diesel source other than on-road vehicles? Some clarification of what is meant by fuel oil here would be helpful.

Author’s response:

As suggested, we have re-written this paragraph in the revised manuscript. We have separately identified potential sources of “gasoline” and “diesel” engines to three sized PM by PCA results based on their fingerprinting elements. (lines 5-25 on page 15, lines 1-14 on page 16 and Table 3 on page 35). In submicron PM, high loadings were found for Ni and V in PC 4. Previous study suggested that Ni and V were attributed to fuel oil combustion from both gasoline and diesel engines (Wang et al., 2003; Shafer et al., 2008), but higher emission rates were for gasoline engines (Cheng et al., 2010), thus this factor is considered as fuel foil combustion from gasoline engines. On the other hand, we agree the reviewer’s comment that better explanations will be given for PCA results if we know the fraction for each traveling traffic type inside the tunnel. Unfortunately, the traffic types in the tunnel provided by Taiwan Area National Freeway Bureau were only divided into LDV and HDV (Table 1); thus we could not know how many fraction of diesel vehicle is.

11th comment

Section 3.5: See my comments above on concerns with the calculation of emission factors in this study. In particular, the use of correction factors of 1.43 and 1.75 for the PM1-1.8 and PM1.8-10 size fractions, respectively, appears to be erroneous. Unless the authors can justify the appropriateness of Eq. 2 for the calculation of emission factors, this section should be removed from the manuscript.

Author's response:

As suggested, we have removed the section 3.5 in the revised manuscript.

12th comment

Figure 1: In the top and middle panels, I found the similarity of colors used made the figures difficult to read. I suggest changing the color scheme in these panels to one with a greater degree of contrast.

Author's response:

As suggested, the top and middle panels in Figure 1 have been re-plotted.

13th comment

Figure 5: This figure does not seem to add much to the manuscript beyond what is already presented in Tables 2 and 5. Suggest removing or moving to Supplemental Information.

Author's response:

Thanks for the reviewer's comment. In the revised manuscript, we have re-organized the second paragraph of section 3.4 (lines 19-25 on page 18 and line 1 on page 19) and all the information of this description are shown in Figure 5. Thus, we have retained Figure 5 in the revised manuscript.

14th comment

Figure 6: See above comments on concerns with emission factor calculations.

Author's response:

The response of this comment is as the same as the 7th comment. In the revised manuscript, we have also removed Figures 6 and 7 which show the results of EmFs for PM metals.

15th comment

P. 13965, line 5: change “impactor” to “impactors”

Author’s response:

We have changed “…deposited impactor (MOUDI)” to “….deposited impactors (MOUDI). (line 6 on page 2)

16th comment

P. 13966, lines 2, 11: change “particulate matters” to “particulate matter”

Author’s response:

As suggested, we have changed “particulate matters” to “particulate matter” in the revised manuscript. (lines 4 and 13 on page 3)

17th comment

P. 13966, lines 12, 17: suggest changing “coarser” to “larger diameter” here, or provide specific ranges of particle diameters

Author’s response:

In the revised manuscript, “coarser particle” has been replaced by “larger particle”. (lines 15 and 19 on page 3)

18th comment

P. 13966 line 24: begin new paragraph with “A number of: : :”

Author’s response:

As suggested, we have created a new paragraph with “A number of …..” on page 4.

19th comment

P. 13967, line 7-8: suggest deleting “..and their associated compositions” as this has already been stated previously in the sentence.

Author’s response:

As suggested, we have deleted “and their associated compositions” in this sentence.

20th comment

P. 13972, line 9: change “peak” to “pick”

Author’s response:

We have corrected “peak” to “pick”. (line 11 on page 10)

21th comment

P. 13981, line 23: change “affect” to “effect”

Author’s response:

Thanks the reviewer’s comment. In the revised, we have deleted the descriptions related EmF and of course, the word of “affect” has also been removed.