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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characteristics of trace metals in traffic-derived particles in Hsuehshan Tunnel, Taiwan: size distribution, fingerprinting metal ratio, and emission factor" *by* Y.-C. Lin et al.

Anonymous Referee #1

Received and published: 25 July 2014

Manuscript ID: acpd-14-13963-2014 Title: Characteristics of trace metals in traffic-derived particles in Hsuehshan Tunnel, Taiwan: size distribution, fingerprinting metal ratio, and emission factor Authors: Y.-C. Lin, C.-J. Tsai, Y.-C. Wu, R. Zhang, K.-H. Chi, Y.-T. Huang, S.-H. Lin, and S.-C. Hsu

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.

Specific Comments:

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.

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.)

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-

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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 equation 2 (see comment on Section 2.4 below).

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?

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

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

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.

Page 13972, lines 1-2: The argument that enhancement of carbonaceous material

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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.

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: C_o/C_i =(EmF×N×L)/(V_air×C_i)+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.

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.

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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.

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.

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.

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.

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

Technical Corrections:

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

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

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

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

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

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

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

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