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### Original article

## Diesel particulate matter and coal dust from trains in the Columbia River Gorge, Washington State, USA



Daniel Jaffe <sup>a, b, \*</sup>, Justin Putz <sup>a</sup>, Greg Hof <sup>a</sup>, Gordon Hof <sup>c</sup>, Jonathan Hee <sup>a</sup>, Dee Ann Lommers-Johnson <sup>a</sup>, Francisco Gabela <sup>a</sup>, Juliane L. Fry <sup>d</sup>, Benjamin Ayres <sup>d</sup>, Makoto Kelp <sup>d</sup>, Madison Minsk <sup>e</sup>

- <sup>a</sup> University of Washington Bothell, School of STEM, Bothell, WA, USA
- <sup>b</sup> University of Washington Seattle, Department of Atmospheric Sciences, Seattle, WA, USA
- <sup>c</sup> Gumbo Software Incorporated, Seattle, WA, USA
- <sup>d</sup> Reed College, Department of Chemistry, Portland, OR, USA
- e Tesla STEM High School, Redmond, WA, USA

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### ABSTRACT

We examined the emissions of diesel particulate matter (DPM) and coal dust from trains in the Columbia River Gorge (CRG) in Washington State by measuring PM<sub>1</sub>, PM<sub>2.5</sub>, CO<sub>2</sub>, and black carbon (BC) during the summer of 2014. We also used video cameras to identify the train type and speed.

During the two-month period, we identified 293 freight trains and 74 coal trains that gave a  $PM_{2.5}$  enhancement of more than 3.0  $\mu g/m^3$ . We found an average  $PM_{2.5}$  enhancements of 8.8 and 16.7  $\mu g/m^3$ , respectively, for freight and coal trains. For most freight trains (52%), and a smaller fraction of coal trains (11%), we found a good correlation between  $PM_{2.5}$  and  $CO_2$ . Using this correlation, we calculated a mean DPM emission factor (EF) of 1.2 gm/kg fuel consumed, with an uncertainty of 20%.

For four coal trains, the videos revealed large plumes of coal dust emanating from the uncovered coal cars. These trains also had the highest peak PM<sub>2.5</sub> concentrations recorded during our study (53–232 µg/m³). Trains with visible coal dust were observed for 5.4% of all coal trains, but 10.3% when the effective wind speed was greater than 90 km/h. We also found that nearly all coal trains emit coal dust based on (1) statistically higher PM<sub>2.5</sub> enhancements from coal trains compared to freight trains; (2) the fact that most coal trains showed a weak correlation between PM<sub>2.5</sub> and CO<sub>2</sub>, whereas most freight trains showed a strong relationship; (3) a statistically lower BC/PM<sub>2.5</sub> enhancement ratio for coal trains compared to freight trains; and (4) a statistically lower PM<sub>1</sub>/PM<sub>2.5</sub> enhancement ratio for coal trains compared to freight trains. Our results demonstrate that, on average, passage of a diesel powered open-top coal train result in nearly twice as much respirable PM<sub>2.5</sub> compared to passage of a diesel-powered freight train. Copyright © 2015 Turkish National Committee for Air Pollution Research and Control. Production and hosting by Elsevier B.V. All rights reserved.

### 1. Introduction

Rail locomotives powered by diesel fuel travel through the Columbia River Gorge National Scenic Area as well as many urban areas in Washington State. Evaluating the air quality impacts from rail traffic for people living near rail lines is hampered by a lack of

E-mail address: djaffe@uw.edu (D. Jaffe).

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data. Several plans that would expand coal shipments by rail through Washington and Oregon to coastal ports for export to Asia have been proposed. New export facilities have been proposed for Longview and Bellingham, Washington. One proposed port near Bellingham would have the capacity to ship up to 54 million metric tons of coal annually (WA DOE, 2013).

The U.S. Department of Health and Human Services states that diesel particulate matter (DPM) is "reasonably anticipated to be a human carcinogen" (U.S. DHHS, 2014). The World Health Organization also categorizes DPM as "carcinogenic to humans" (WHO, 2012). In urban areas, including Seattle, the most significant "air toxic" is DPM, contributing over 80% of the cancer risk for air toxics

<sup>\*</sup> Corresponding author. University of Washington Bothell, School of STEM, Bothell, WA, USA.

(Keill and Maykut, 2003; PSCAA, 2005). DPM sources consist of rail locomotives, ships and diesel trucks, both on road and off road. Average DPM concentrations for the Seattle area are  $1.4-1.9 \,\mu g/m^3$ , based on monitoring and a chemical mass balance model (Keill and Maykut, 2003; Maykut et al., 2003). These DPM concentrations make up 15-20% of the mass of total particulate matter with diameters less than  $2.5 \,\mu m$  (PM<sub>2.5</sub>).

Emission standards for new and remanufactured locomotives, developed by the U.S. Environmental Protection Agency (EPA) (40 CFR part 1033) have decreased steadily over the past several decades. For diesel locomotives various standards apply based on the date of manufacture: Tier 0, 1973–2001; Tier 1, 2002–2004; Tier 2, 2005–2010; Tier 3, 2011–2014; and Tier 4, after 2015 (U.S. EPA, 2013). Tier 4 locomotives must comply with a PM<sub>10</sub> standard of 0.03 g/bhp-h, which is about 0.19 g of PM<sub>10</sub> per kg of fuel consumed (U.S. EPA, 2009).

Previous studies looked at rail yards as air pollutant sources. They determined that the primary source of PM<sub>2.5</sub> at these sites was diesel fuel combustion. One study investigated the impact of DPM emissions on PM2.5 concentrations at an Atlanta area rail yard (Galvis et al., 2013). Using measurements collected upwind and downwind of the rail yard, they found the average "neighborhood" contribution to PM<sub>2.5</sub> was 1.7  $\mu$ g/m<sup>3</sup>. The emission factors (EFs) per kg of diesel fuel burned were calculated to be 0.4-2.3 g DPM. The EFs were not determined from individual train measurements but were calculated using three different methods, each based on differing assumptions. Two studies of a Roseville, California, rail yard also found significant enhancements in PM25 from the yard. Using measurements from upwind and downwind, Cahill et al. (2011) found an average PM<sub>2.5</sub> enhancement of 4.6 μg/m<sup>3</sup>, and Campbell and Fujita (2006) found even larger contributions (7.2–12.2 µg/ m<sup>3</sup>). Cahill et al. (2011) also demonstrated that particles with diameters below 1 μm are the major contributor to PM<sub>2.5</sub> aerosol mass from diesel exhaust. Abbasi et al. (2013) studied concentrations in the interior of trains and close to rail lines and found significantly elevated PM<sub>2.5</sub> and PM<sub>10</sub> concentrations, particularly in stations that were underground. Gehrig et al. (2007) looked at electric trains in Switzerland and examined the influence of dust from these trains on PM<sub>10</sub> concentrations. Several studies investigated the EFs of on-road diesel trucks and buses (Jamriska et al., 2004; Zhu et al., 2005; Cheng et al., 2006; Park et al., 2011; Dallmann et al., 2012), but we have found no similar studies on diesel rail.

Trains that carry coal in uncovered rail cars may also release coal dust, in addition to DPM, into the atmosphere. The BNSF railway requires that a surfactant be applied over the top of coal being transported by rail (see BNSF Railway, 2013). However, we are unaware of any studies reported in the scientific literature that evaluate the efficacy of this or the impact of coal dust on air quality. By examining the PM by train type, we can examine whether there is respirable coal dust (PM<sub>2.5</sub>) as part of the emissions from coal trains. We will also examine the particle size distribution because combustion-related particles and coal dust, which is mechanically generated, are associated with particles of different sizes (Seinfeld, 1986)

A substantial amount (44–60%) of the diesel engine PM<sub>2.5</sub> mass is black carbon (BC) (Bond et al., 2004; Kirchstetter and Novakov, 2007; Ramanathan and Carmichael, 2008). Because radiative forcing due to BC is the major light-absorbing species in atmospheric aerosol, it is significant both globally and regionally (Jacobson, 2001; Ramanathan and Carmichael, 2008). In addition, because of BC's surface properties, it is possible for polyaromatic hydrocarbons (PAHs) and other semi-volatile compounds to be adsorbed and transported by BC (Dachs and Eisenreich, 2000). Health organizations are also taking a hard look at BC because of its contribution to the harmful effects caused by PM<sub>2.5</sub>, including cardiopulmonary

and respiratory disease (Jansen et al., 2005; Janssen et al., 2011; U.S. EPA, 2012).

Because of the lack of information on  $PM_{2.5}$  concentrations and the exposure to humans from diesel trains, the debate over coal dust and the scarcity of information on diesel train EFs, we sought to measure these air quality effects by answering the following questions:

- 1. What are the DPM emission factors for locomotives in Washington State and how do these compare with published values?
- 2. Do open-top coal-carrying trains emit respirable coal dust (PM<sub>2.5</sub>) into the air? If so, can we quantify the emissions?

To address these questions we measured PM<sub>1</sub>, PM<sub>2.5</sub>, CO<sub>2</sub>, black carbon and meteorology at a location in the Columbia River Gorge next to the rail line. Because we wanted to quantify DPM and coal dust exposure and quantify the EFs from each train, we collected measurements every 10 s in order to identify the air quality impacts of individual trains. In a previous study, we measured a similar suite of parameters in 2013 at a site in Seattle, Washington, and (very briefly) at a site in the Columbia River Gorge (Jaffe et al., 2014). In the previous study, we quantified DPM emission factors from diesel trains, evaluated the neighborhood scale exposure to PM<sub>2.5</sub> from trains and found evidence that suggested emissions of coal dust, based on particle size. In the present analysis, we report new data taken in 2014 that more clearly identifies and quantifies the emissions of DPM and coal dust from coal-carrying trains.

### 2. Experimental

Measurements were made at a site between the towns of Lyle and Dallesport, Washington, in the Columbia River Gorge (approximately 45.7°N, 121.2°W) between June 7—August 10, 2014. The instruments were housed in a weather-proof enclosure, located about 10 m above and 20 m northeast of the rail line. Two video cameras were used; one took video of the trains at a 90° angle to the rail line, and one viewed the trains arriving/departing to the northwest. The rail line travels along the north side of the Columbia River. There were no roads between our site and the river. Our measurement site was approximately 200 m southwest of Washington Route 14, a state highway with light traffic. The measurement location used in 2014 was in the same general location, but about 300 m away, from the site we used for our 2013 measurements (Jaffe et al., 2014). At this site the rail line is almost completely flat; there is a maximum grade of 1 m per km in the next few km in either direction.

We used a DustTrak DRX Aerosol Monitor (Model #8533, TSI, Inc., Shoreview, MN) to measure size-segregated PM. The DustTrak reports 4 size fractions of PM mass concentrations: PM<sub>1</sub>, PM<sub>2.5</sub>, PM<sub>10</sub> and TSP. The instrument uses aerosol scattering to calculate its measurements. Therefore, its measurements are not the same as mass-based measurements (Wang et al., 2009). The DustTrak is calibrated against Arizona road dust (ISO 12103-1) by the manufacturer and so will not correctly reflect the mass concentration for other types of aerosol. This is specifically the case for diesel PM because of the particle size (Park et al., 2011). Obtaining accurate measurements with the DustTrak requires comparing its measurements with a mass-based measurement (Moosmuller et al., 2001). The DustTrak has been used to quickly measure several PM size fractions and determine EFs of individual vehicles in several previous studies (e.g., Park et al., 2011; Dallmann et al., 2012), but usually after using a mass-based method to calibrate the response factor (Jamriska et al., 2004; Zhu et al., 2005; Cheng et al., 2006; Jaffe et al 2014). In our study, the DustTrak was calibrated against two mass-based measurements—a Tapered Element Oscillating Microbalance (TEOM) and the EPA Federal Reference Method at a routine air quality monitoring station in Seattle, Washington (details below).

The DustTrak inlet was stainless steel tubing (4.8 mm i.d.) facing downward from a height of approximately 2 m above ground level. The flow rate through the inlet was 3.0 L per minute. With these conditions, the flow was laminar. To estimate the particle sampling efficiency, we used the methodology and program provided by von der Weiden et al. (2009). The wind speeds during train sampling in the CRG varied between 1 and 11 m per second (mps), with an average of 4.5 mps during the sampling period. For particles less than 2.5  $\mu m$  aerodynamic diameter, we calculated greater than 90% particle transmissions at all wind speeds up to 15 mps. For particles between 3 and 10  $\mu m$  aerodynamic diameter, the inlet sampling efficiency would be much less than 1.0 and vary with wind speed (von der Weiden et al., 2009). For this reason, we used only the PM<sub>2.5</sub> and PM<sub>1</sub> data in this analysis.

We measured CO<sub>2</sub> using a Licor-820 (Licor, Inc., Lincoln, NE) with a small vacuum pump for sampling. The inlet was a 4.8 mm i.d. stainless steel tube (38 mm long) connected to PFA tubing. We zeroed the instrument using CO<sub>2</sub>-free air and calibrated it with a 395 ppmv standard from Airgas, Inc. We calibrated the instrument both before and after the deployment; the instrument response varied by less than 1 ppmv between these calibrations. We used DAQFactory on a PC to record data from the DustTrak, the Licor-820 (CO<sub>2</sub>, cell temperature and pressure) and the meteorological station. We recorded 10-s averages for PM and CO<sub>2</sub> data.

To identify trains and quantify their speeds, we used two Night Owl cameras (Model CAM-MZ420-425M) that were equipped with infrared (IR) night vision. The cameras were motion activated and operated with iSpy open source camera security software. However, even with the IR capability of the cameras, we were unable to identify the type of trains at night. We considered using an auxiliary light to view the trains at night; however, this was rejected as the Columbia River Gorge is classified as a National Scenic Area, which limits lighting options. Only trains that could positively be identified as freight or coal were used in this analysis, so this excluded all trains passing our site in full darkness.

BC was measured using an aethalometer (Magee Scientific model AE22). BC data were collected at one-minute time resolution at 370 nm and 880 nm. BC loading was determined using infrared attenuation data at 880 nm alone, because at 370 nm, other organic compounds may contribute interference (Wang et al., 2011). The aethalometer determines raw BC concentration (BC0, ng/m³) from measured attenuation values (ATN,  $m^{-1}$ ) via

$$BC_0 = 10^9 \times ATN/\sigma \tag{1}$$

where  $\sigma$  is the calibrated cross-section (16.6  $m^2/g$  at 880 nm). As in our previous study (Jaffe et al., 2014), we applied a correction to the BC $_0$  concentrations to account for diminishing transmission as a function of BC loading. Transmission (Tr) is calculated from each attenuation value:

$$Tr = e^{-ATN/100} \tag{2}$$

Following Kirchstetter and Novakov (2007), we calculated the corrected BC mass loading ( $BC_{corr}$ ,  $ng/m^3$ ) as:

$$BC_{corr} = BC_0/(0.88 \times Tr + 0.12)$$
 (3)

The DPM EFs are calculated for each passing train in units of DPM emitted per kg of diesel fuel burned using:

$$EF\left(PM_{2.5}\right) = \frac{\Delta PM_{2.5}}{\Delta CO_{2}} \times CF \times W_{c} \tag{4} \label{eq:effective}$$

where the  $\Delta PM_{2.5}/\Delta CO_2$  or "enhancement ratio" is calculated from the Reduced Major Axis (RMA) regression slopes of the 10-s  $CO_2$  and  $PM_{2.5}$  data for each passing train, in units of  $\mu g/m^3$  per ppmv. CF is a conversion factor to convert  $CO_2$  concentrations in ppm to  $\mu g$   $C/m^3$  units using the ideal gas law at 1 atm and 25 °C (1 ppmv  $CO_2 = 490.7$  ugC/m³).  $W_C$  is the mass fraction of carbon in diesel fuel (870 g C/kg fuel) (Lloyd's Register, 1995; Cooper, 2003), which yields overall units on the EF of g  $PM_{2.5}/kg$  fuel consumed. Yanowitz et al. (2000) showed that over 95% of diesel fuel carbon is released as  $CO_2$ .

Enhancement ratios ( $\Delta PM_{2.5}/\Delta CO_2$  and  $\Delta PM_1/\Delta PM_{2.5}$ ) were calculated from the 10-s data using the RMA regression method, which considers errors in both the x and y variables (Ayers, 2001; Cantrell, 2008). Absolute enhancements were calculated by subtracting out the PM, BC and  $CO_2$  maximums during train passage from the background concentration measured prior to each trains passage. The RMA regression parameters were calculated for each train passage using a program written in Java utilizing Apache Commons Mathematics Library 3.3. The program first looked for a PM<sub>2.5</sub> enhancement of at least 3  $\mu$ g/m³ over the median value from the past 17 min (100, 10-s data points). The accuracy of the Java program to calculate PM and  $CO_2$  enhancements and the RMA regression parameters were manually verified for approximately 20% of the peaks. All times in this manuscript are given in Pacific Daylight Time (PDT).

### 3. Results

### 3.1. Calibration of the DustTrak

We compared the DustTrak PM<sub>2.5</sub> concentrations with a TEOM and the filter-based Federal Reference Method (FRM) at a routine air quality monitoring site in Seattle, Washington (Beacon Hill), operated by the Puget Sound Clean Air Agency (PSCAA). Comparison data were obtained between April 30—May 20, 2014. TEOM data were continuous and reported on an hourly basis, the filter-based FRM measurements were for 24 h and conducted every third day only. At this site, the TEOM is a Thermo Fisher Scientific Model 1400AB with 8500C Filter Dynamic Measurement System (FDMS) with the Very Sharp Cut Cyclone (VSCC<sup>TM</sup>) modification (U.S. EPA, 2014). This configuration is designated by the EPA as a Federally Equivalent Method (FEM) for PM<sub>2.5</sub>. The inlet and flow configuration used for the DustTrak at the Beacon Hill site were identical to the configuration used in the Columbia River Gorge.

We found a very good correlations between the TEOM  $PM_{2.5}$ , the FRM and the DustTrak's reported  $PM_{2.5}$ . Table 1 shows the regression parameters.

The 95% confidence interval in the slope for the DustTrak-TEOM comparison is  $\pm 4.5\%$ , whereas it is  $\pm 32\%$  for the DustTrak-FRM comparison due to the very small sample size. In both cases, the intercepts are insignificantly different from zero (95% confidence interval overlaps zero). Because of this, we corrected all of the DustTrak PM data using the TEOM slope of 0.5577. This slope is 22% greater than the one reported by Jamriska et al. (2004), who reported a slope of 0.458. It also is approximately 14% greater than our earlier DustTrak comparison at a different site, where we reported a slope of 0.491 (Jaffe et al., 2014). These differences may be attributable to different aerosol types at these sites. Given these differences, we estimated the uncertainty in the corrected DustTrak PM1 and PM2.5 values to be  $\pm 20\%$ .

**Table 1**Regression parameters for the comparisons between the DustTrak data, the TEOM data and the FRM method at the PSCAA site at Beacon Hill, Seattle, Washington.

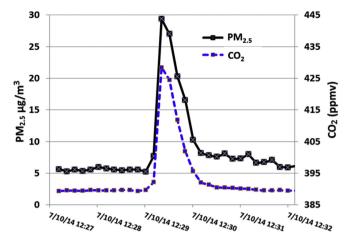
Comparison equation (using reduced major axis regression)	R <sup>2</sup>	N
TEOM PM <sub>2.5</sub> ( $\mu$ g/m <sup>3</sup> ) = DustTrak × 0.5577 $-$ 0.6977 FRM PM <sub>2.5</sub> = DustTrak × 0.5524 $-$ 0.8433 FRM PM <sub>2.5</sub> = TEOM × 1.05 $-$ 0.4326	0.92	485 (h averages) 7 (24-h samples) 7 (24-h samples)

# 3.2. Overview of observations on train emissions in the Columbia River Gorge

As each train passed our observation site, we may detect a peak in PM and CO<sub>2</sub>, but this depended on the wind direction and wind speed. If the winds were from the north to northeast directions, our sensors recorded minor peaks only, or no peaks at all, in PM and CO<sub>2</sub>. We found that small PM events had a lower correlation between the various parameters. For this reason, we screened out small peaks where the maximum  $\Delta PM_{2.5}$  (enhancement above background) was  $<3 \mu g/m^3$ . If a peak larger than this value was detected and the video confirmed a simultaneous train passage, then we included this peak in our analysis. We included only freight and coal-carrying trains, since these were the dominant types that we observed in the Columbia River Gorge. Trains that carried mixed loads (e.g., freight plus coal), sand or other unidentifiable or uncovered cargo were not included in this analysis. We also observed very few passenger trains during the daytime hours, in contrast to our previous study in Seattle (Jaffe et al., 2014).

During this study, we observed 367 events with  $\Delta PM_{2.5} > 3 \mu g/s$ m<sup>3</sup> that were identified by the video cameras as either freight or coal. We refer to each train passage with a detectable PM peak and verified by the video as a "train event." Table 2 shows a summary of the 367 train events, including number and average peak PM<sub>1</sub> and PM<sub>2.5</sub> enhancement values (over background). The peak PM<sub>1</sub> and PM<sub>2.5</sub> enhancements (10-s) from coal trains are about double the enhancements seen from freight trains. In addition, there are three extreme events with PM<sub>2,5</sub> enhancements greater than 75 μg/m<sup>3</sup> that were seen only for the coal trains. The differences between the peak PM enhancements for coal and freight trains are statistically significant (P < .001). The statistically significant difference remains even if these extreme events are excluded from the analysis. For all train events, there is an excellent relationship between the PM<sub>1</sub> and PM<sub>2.5</sub> data, although the fraction of PM<sub>1</sub>/PM<sub>2.5</sub> varies by train type. This is discussed in Section 3.5 below.

However, only some train events showed a good correlation between  $PM_{2.5}$  and  $CO_2$ . Fig. 1 shows an example of a freight train that passed our site on July 10, 2014. In this case, the  $PM_{2.5}$  enhancement is  $24 \mu g/m^3$ , the  $CO_2$  enhancement is 39 ppmv and the two are very well correlated, indicating that the dominant source of PM is diesel exhaust. Fig. 2 shows an example of a coalcarrying train that passed by on July 18, 2014. For this example, the peak  $PM_{2.5}$  concentration is more than 6 times the peak shown



**Fig. 1.** PM<sub>2.5</sub> and CO<sub>2</sub> during passage of a freight train on 7/10/2014 at 12:29 PDT. The two values show a good correlation with an R<sup>2</sup> of 0.98 and a slope of 0.61  $\mu$ g/m<sup>3</sup> per ppmv.

previously for the freight train, while the CO<sub>2</sub> enhancement is much smaller. In addition, the CO<sub>2</sub> peaks occurred at the start and end of the train passage due to locomotives at the beginning and end of this train, which is typical of the very long coal trains. The height of the CO<sub>2</sub> peak shows no obvious relationship with train type and likely varies mainly with meteorology, which influences the degree to which the combustion exhaust gases reach the measurement site. For the coal train (Fig. 2), the dominant source of PM is not diesel exhaust but coal dust. This was confirmed by the video (discussed below). It should be noted that DPM was probably present but is not apparent in the data due to the much larger coal dust peak. In this case, because the PM concentrations were not correlated to CO<sub>2</sub>, we were not able to calculate a DPM emission factor. For this reason, we did not include train events in the DPM EF calculation if the PM<sub>2.5</sub>-CO<sub>2</sub> R<sup>2</sup> is less than 0.5. We also excluded train events that had very small  $CO_2$  enhancements ( $\Delta CO_2 < 2$ ppmv), as these had erratic behavior.

Supplementary data related to this article can be found online at http://dx.doi.org/10.1016/j.apr.2015.04.004

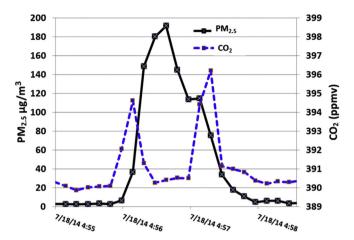
### 3.3. DPM emission factors

The  $\Delta PM_{2.5}/\Delta CO_2$  was used to derive the DPM emission factors. The average  $\Delta PM_{2.5}/\Delta CO_2$  slope for all train events was found to be 6.56  $\mu g/m^3$  per ppmv, but this included many trains with a very poor correlation between  $PM_{2.5}$  and  $CO_2$ . For the DPM emission factor calculation, we restricted our analysis to only those cases with an  $R^2$  for the  $PM_{2.5}-CO_2$  relationship of 0.5 or greater and a  $CO_2$  enhancement of at least 2 ppmv. Table 2 shows the number of each train type that was used for the DPM analysis and statistics on the  $PM_{2.5}-CO_2$  slope.

Table 2 PM and CO<sub>2</sub> data for freight and coal trains. Slopes for  $\Delta$ PM<sub>2.5</sub>/ $\Delta$ CO<sub>2</sub> relationship is reported only for those train events with R<sup>2</sup> >0.5 and  $\Delta$ CO<sub>2</sub> >2 ppmv.<sup>a</sup>

	Freight	Coal	All trains
Number	293	74	367
Average peak $\Delta PM_1$ (µg/m <sup>3</sup> )	11.0	19.7	12.5
Average peak $\Delta PM_{2.5}$ (µg/m <sup>3</sup> )	10.7	20.9	13.0
Maximum $\Delta PM_{2.5} (\mu g/m^3)$	57.2	232.3	232.3
Number with $PM_{2.5} - CO_2 R^2 > 0.5$ and $\Delta CO_2 > 2$ ppm	152 (52%)	11 (15%)	163 (44%)
Mean/median ΔPM <sub>2.5</sub> /ΔCO <sub>2</sub> slope (µg/m <sup>3</sup> /ppmv)	0.70/0.56	0.71/0.56	0.70/.56
Max/Min slope	3.88/0.10	1.64/0.20	3.88/0.10

a In addition to the criteria given in the text above, we excluded one additional case with visible coal dust and an extremely high PM2.5—CO2 slope (12.0).



**Fig. 2.** PM $_{2.5}$  and CO $_2$  during passage of a coal train on 7/18/2014 at 4:56 PDT. The two parameters show no correlation during this time period. The train was observed to have locomotives in the front and rear, giving rise to the CO $_2$  peaks at the beginning and end of this time period.

The data in Table 2 show that while most freight trains were included in this analysis, the majority of coal trains were not included. This is due to the fact that most of the coal train events show a poor correlation between PM<sub>2.5</sub> and CO<sub>2</sub> (see Fig. 2). One coal train that would otherwise have been included in the DPM calculation had a PM<sub>2.5</sub> — CO<sub>2</sub> slope of 12.0, more than  $10\times$  the mean value, and had visible coal dust in the video. Thus the large amount of PM<sub>2.5</sub> in this case cannot be attributed solely to DPM. This train event was not included in the DPM analysis. With this exclusion, the mean and median slopes for freight and coal trains are rather similar. Using equation (4), we find that the mean and median DPM EFs from our study are 1.2 and 0.99 g/kg fuel consumed, with an overall uncertainty of 20%. Our previous observations in the Pacific Northwest (Jaffe et al., 2014) found an average EF for diesel locomotives of 0.94 g/kg.

Diesel EFs for locomotives have been previously reported from several measurement campaigns. Kean et al. (2000) reported locomotive emission factors of between 1.8 and 2.1 g/kg using the EPA "NONROAD" model. A 2009 report (U.S. EPA, 2009) estimated that average locomotives EFs are declining about 5% per year, with a 2014 value of 0.98 g/kg. A study by Sierra Research in 2004 (Sierra Research, 2004) forecast a much slower decrease in the EFs of diesel locomotives, compared to U.S. EPA (2009), and for 2014 projected 1.4 g/kg. Our average measured EF is consistent with those cited in the above literature for the 2014 time frame, within the respective uncertainties.

### 3.4. Black carbon

We obtained simultaneous BC and  $PM_{2.5}$  data on 294 of the trains. Table 3 reports the observed  $BC/PM_{2.5}$  and  $PM_1/PM_{2.5}$  enhancement ratios (discussed in Section 3.5).

These data show that, on average, 43% of the  $PM_{2.5}$  was BC for all trains. In our previous study using similar data from 2013 (Jaffe et al., 2014), we found that the  $BC/PM_1$  fraction was 52%, with most of those observations on freight trains. Our new data in 2014 indicates a significant difference (P < .001) in the average  $BC/PM_{2.5}$  fraction for freight (0.47) and coal trains (0.29). Previous studies have found values that are similar to our freight train values for the BC/PM fraction. A study by Hildemann et al. (1991) found that 55% of diesel emissions were BC, and Watson et al. (1994) reported 45%. An Atlanta study (Galvis et al., 2013) found that diesel trains had BC to  $PM_{2.5}$  ratios of 47–52%. The significant difference in the  $BC/PM_{2.5}$ 

**Table 3** BC/PM<sub>2.5</sub> and PM<sub>1</sub>/PM<sub>2.5</sub> enhancement ratios for freight and coal trains.

	Freight	Coal	All trains
N (for BC/PM <sub>2.5</sub> analysis)	233	61	294
Mean/median BC/PM <sub>2.5</sub> (unitless)	0.47/0.40	0.29/0.20	0.43/0.35/0.27
Standard deviation on BC/PM <sub>2.5</sub>	0.27	0.23	0.27
N (for PM <sub>1</sub> /PM <sub>2.5</sub> analysis)	293	74	367
Mean/median PM <sub>1</sub> /PM <sub>2.5</sub> (unitless)	0.93/0.93	0.96/0.96	0.96/0.96
Standard deviation on PM <sub>1</sub> /PM <sub>2.5</sub>	0.03	0.03	0.03

between coal and freight trains, shown in Table 3, indicates a significant coal dust component in the PM from the coal trains.

We assume that the coal dust has the same composition as the coal being shipped. This coal, from the Powder River Basin of Wyoming and Montana, has a relatively low carbon content compared to other coal types (ca 50% C), with the remainder of the mass made up of moisture and minerals, such as silicates, iron oxides and calcium oxide (NETL, 2012). While the low carbon content is partly responsible for the low BC/PM<sub>2.5</sub> fraction, shown in Table 3, our data suggest that other factors may also be involved. This could include a change in the mass absorption cross section for coal dust, as compared to diesel exhaust, which might reflect the impact of the coal mineral content, the organic matter composition or the size distribution of the particles.

### 3.5. $PM_1/PM_{2.5}$ fraction

The DustTrak calculates concentrations of PM in four size ranges, but due to the inlet sampling efficiency (discussed in Section 2) we considered only data for PM $_1$  and PM $_2$ .5. Table 3 gives the statistical parameters on the PM $_1$ /PM $_2$ .5 enhancement ratio. Coal trains showed a larger mass fraction of particles above 1  $\mu$ m aerodynamic diameter, and this difference is statistically significant. This reflects the significant contribution of coal dust to the PM $_2$ .5 concentrations during the passage of the coal trains.

### 3.6. Influence of coal dust on PM<sub>2.5</sub> concentrations

In four cases, the videos revealed visible coal dust from the open-top coal trains. These visible coal dust plumes were seen in the four train events with the highest peak  $PM_{2.5}$  concentrations (Table 4). We call these four train events with the highest  $PM_{2.5}$  and visible coal dust "super-dusters." Two of the "super-duster" videos have been archived as part of the supplemental materials for this paper (8/7/2014 and 7/27/2014). Fig. 3 shows still images obtained from the video before and after train passage for the "super duster" on 8/7/2014, along with the measured  $PM_{2.5}$  concentrations. We found that 4 out of 74 coal trains, or 5.4%, were classified as "super dusters" during our study.

A number of factors could be important in explaining the coal dust emissions of PM<sub>2.5</sub> from coal trains. These include quality of the surfactant application or factors that may disturb the coal/surfactant surface, such as high train speeds, exposure to high winds or rough handling during transport. While we have no information on

**Table 4** The four train events with the highest peak  $PM_{2.5}$  concentrations. In each case, a coal train with a visible coal dust plume was confirmed in the video recording.

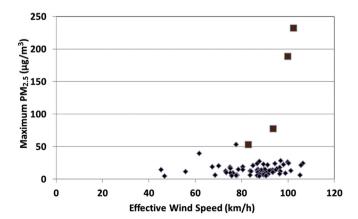
Date/time (PDT)	Peak PM <sub>2.5</sub> conc. μg/m <sup>3</sup>	Peak BC μg/m <sup>3</sup>	BC/PM <sub>2.5</sub> ratio
8/7/14 17:28	232.3	53.5	0.23
7/18/14 4:57	188.8	88.9	0.47
7/20/14 14:07	77.6	8.86	0.11
7/27/14 21:16	53.1	9.13	0.17



**Fig. 3.** Images captured from the video camera before and after coal train passage on 8/7/2014 at 17:28 PDT. The full video of this train passage is archived as part of the supplemental materials for this paper. The camera looks to the west, downriver in the Columbia River Gorge. The coal train is visible in the right image and was moving from left to right.

upstream conditions, our data do allow us to examine the influence that train and local wind speed may have played on dust emissions. To do this, we calculated train speeds for each coal train from the videos. We also calculated the vector component of the winds in the direction opposite to the trains' travel. The sum of train speed plus vector wind speed represents the true wind speed across the opentop coal trains. We refer to this as the effective wind speed. During our study, the average train speed was 71.3 km/h and the average vector wind speed was 14.9 km/h.

Fig. 4 shows the effective wind speed versus peak PM<sub>2.5</sub> for each coal train event. The four "super dusters" are shown as large red squares. While no simple relationship emerges from this analysis, the data do suggest that "super dusters" are more likely to occur when the effective wind speed is greater than 80–90 km/h. Above 90 km/h, the fraction of "super dusters" is 10.3% (3 out of 29 trains), compared to 5.4% at all wind speeds. Thus we can view wind speed as one factor that increases the risk of high-level coal dust exposure. However, the fact that many coal trains with effective wind speeds greater than 90 km/h are not "super dusters" indicates that other factors, such as quality of the surfactant applied to the coal surface, must also be important.



**Fig. 4.** Peak PM $_{2.5}$  enhancement for each coal train passage versus effective wind speed over the top of the train. The effective wind speed is calculated as the train speed plus the vector component of the wind at  $180^{\circ}$  to the train's movement. The four "super dusters" are shown as large red squares.

### 4. Conclusions

We measured PM<sub>1</sub>, PM<sub>2.5</sub>, BC and CO<sub>2</sub> during 367 train passages (train events) in the Columbia River Gorge. From the data, we calculated a DPM EF average of 1.2 g/kg fuel consumed ( $\pm 20\%$ ) on 163 of those train events that show a good correlation between PM<sub>2.5</sub> and CO<sub>2</sub> (mostly freight trains). Our data indicate that nearly all open-top coal trains release coal dust, which contributes to enhanced PM<sub>2.5</sub> in the Columbia River Gorge. In four train events, that we call "super-dusters," the coal dust emissions led to visible dust plumes and the highest PM<sub>2.5</sub> concentrations observed in our study. But nearly all coal trains generate some degree of coal dust (PM<sub>2.5</sub>) based on the following evidence:

- 1. Statistically higher peak PM<sub>2.5</sub> concentrations during passage of coal trains compared to freight trains. The peak PM<sub>2.5</sub> enhancements during a coal train passage are nearly double, on average, compared to the value during a freight train passage (Table 2);
- 2. The fact that most freight trains (52%) show a good correlation between PM<sub>2.5</sub> and CO<sub>2</sub>, whereas very few coal trains (15%) show this relationship (Table 2);
- 3. The BC/PM<sub>2.5</sub> enhancement ratio is statistically higher for freight trains compared to coal trains (Table 3);
- 4. The PM<sub>1</sub>/PM<sub>2.5</sub> enhancement ratio is statistically higher during passage of freight trains compared to coal trains (Table 3).

These four results demonstrate statistically significant differences between freight and coal trains, even if the four superdusters are excluded from the statistical analysis.

Because our focus was on air quality, we measured the respirable size fractions of PM. Thus it is not possible to relate our observations to any data on bulk loss of coal during transport, since most of this loss will occur as much larger size particles. Because most coal train events show a poor correlation between PM<sub>2.5</sub> and CO<sub>2</sub>, it is not possible to rigorously derive a fuel-based emission factor for the coal dust. Nonetheless, our data provide some guidance to anyone wishing to calculate total PM<sub>2.5</sub> emissions from the railway sector. Since the peak PM<sub>2.5</sub> values for coal trains are nearly double those for freight trains, it is reasonable to conclude that the total PM<sub>2.5</sub> emissions from coal trains are approximately double

those of freight trains. This would imply that the coal train PM<sub>2.5</sub> emissions consist of approximately half DPM and half coal dust.

Though all coal trains appear to generate some degree of dust, the "super-dusters" generate visible plumes and the highest concentrations of  $PM_{2.5}$ . "Super-dusters" represent 5.4% of all coal trains but 10.3% when the effective wind speed is greater than 90 km/h. This indicates that wind is one factor contributing to the coal dust emissions, but it is not the only explanatory factor.

### **Conflict of interest**

The authors have no conflicts of interest to report.

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### References

- Abbasi, S., Jansson, A., Sellgren, U., Olofsson, U., 2013. Particle emissions from rail traffic: a literature review. Crit. Rev. Environ. Sci. Technol. 43, 2511–2544.
- Ayers, G.P., 2001. Comment on regression analysis of air quality data. Atmos. Environ. 35, 2423–2425.
- BNSF Railway, 2013. BNSF Railway Statement on STB Coal Dust Decision (accessed in 2015). http://www.bnsf.com/customers/what-can-i-ship/coal/coal-dust.html.
- Bond, T.C., Streets, D.G., Yarber, K.F., Nelson, S.M., Woo, J.H., Klimont, Z., 2004. A technology-based global inventory of black and organic carbon emissions from combustion. J. Geophys. Research-Atmos. 109 art. no. D14203.
- Cahill, T.A., Cahill, T.M., Barnes, D.E., Spada, N.J., Miller, R., 2011. Inorganic and organic aerosols downwind of California's Roseville Railyard. Aerosol Sci. Technol. 45, 1049–1059.
- Campbell, D.E., Fujita, E.M., 2006. Data Analysis on the Roseville Rail Yard Air Monitoring Project. Year One Interim Report. http://www.placer.ca.gov/~/media/apc/documents/UP/2006/April/
  - UPRRAMP2005DataAnalysisFinalReport041306.pdf (accessed in 2015).
- Cantrell, C.A., 2008. Technical note: review of methods for linear least-squares fitting of data and application to atmospheric chemistry problems. Atmos. Chem. Phys. 8, 5477–5487.
- Cheng, Y., Lee, S.C., Ho, K.F., Louie, P.K.K., 2006. On-road particulate matter (PM<sub>2.5</sub>) and gaseous emissions in the Shing Mun Tunnel, Hong Kong. Atmos. Environ. 40, 4235–4245.
- Cooper, D.A., 2003. Exhaust emissions from ships at berth. Atmos. Environ. 37, 3817–3830.
- Dachs, J., Eisenreich, S.J., 2000. Adsorption onto aerosol soot carbon dominates gasparticle partitioning of polycyclic aromatic hydrocarbons. Environ. Sci. Technol. 34, 3690–3697.
- Dallmann, T.R., DeMartini, S.J., Kirchstetter, T.W., Herndon, S.C., Onasch, T.B., Wood, E.C., Harley, R.A., 2012. On-road measurement of gas and particle phase pollutant emission factors for individual heavy-duty diesel trucks. Environ. Sci. Technol. 46, 8511–8518.
- Galvis, B., Bergin, M., Russell, A., 2013. Fuel-based fine particulate and black carbon emission factors from a railyard area in Atlanta. J. Air Waste Manag. Assoc. 63, 648–658.
- Gehrig, R., Hill, M., Lienemann, P., Zwicky, C.N., Bukowiecki, N., Weingartner, E., Baltensperger, U., Buchmann, B., 2007. Contribution of railway traffic to local PM<sub>10</sub> concentrations in Switzerland. Atmos. Environ. 41, 923–933.
- Hildemann, L.M., Markowski, G.R., Cass, G.R., 1991. Chemical-composition of emissions from urban sources of fine organic aerosol. Environ. Sci. Technol. 25, 744–759.
- Jacobson, M.Z., 2001. Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. Nature 409, 695–697.
- Jaffe, D.A., Hof, G., Malashanka, S., Putz, J., Thayer, J., Fry, J.L., Ayres, B., Pierce, J.R., 2014. Diesel particulate matter emission factors and air quality implications from in-service rail in Washington State, USA. Atmos. Pollut. Res. 5, 344–351.
- Jamriska, M., Morawska, L., Thomas, S., He, C., 2004. Diesel bus emissions measured in a tunnel study. Environ. Sci. Technol. 38, 6701–6709.

- Jansen, K.L., Larson, T.V., Koenig, J.Q., Mar, T.F., Fields, C., Stewart, J., Lippmann, M., 2005. Associations between health effects and particulate matter and black carbon in subjects with respiratory disease. Environ. Health Perspect. 113, 1741–1746.
- Janssen, N.A.H., Hoek, G., Simic-Lawson, M., Fischer, P., van Bree, L., ten Brink, H., Keuken, M., Atkinson, R.W., Anderson, H.R., Brunekreef, B., 2011. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM<sub>10</sub> and PM<sub>2.5</sub>. Environ. Health Perspect. 119, 1691–1699.
- Kean, A.J., Sawyer, R.F., Harley, R.A., 2000. A fuel-based assessment of off-road diesel engine emissions. J. Air Waste Manag. Assoc. 50, 1929—1939.
- Keill, L., Maykut, N., 2003. Final Report: Puget Sound Air Toxics Evaluation. Puget Sound Clean Air Agency.
- Kirchstetter, T.W., Novakov, T., 2007. Controlled generation of black carbon particles from a diffusion flame and applications in evaluating black carbon measurement methods. Atmos. Environ. 41, 1874–1888.
- Lloyd's Register, 1995. Marine Exhaust Emissions Research Program, London.
- Maykut, N.N., Lewtas, J., Kim, E., Larson, T.V., 2003. Source apportionment of PM<sub>2.5</sub> at an urban IMPROVE site in Seattle, Washington. Environ. Sci. Technol. 37, 5135—5142
- Moosmuller, H., Arnott, W.P., Rogers, C.F., Bowen, J.L., Gillies, J.A., Pierson, W.R., Collins, J.F., Durbin, T.D., Norbeck, J.M., 2001. Time resolved characterization of diesel particulate emissions. 1. Instruments for particle mass measurements. Environ. Sci. Technol. 35, 781–787.
- NETL (National Energy Technology Laboratory), 2012. Detailed Coal Specifications. DOE/NETL-401/012111. http://www.netl.doe.gov/File%20Library/research/energy%20analysis/publications/QGESS\_DetailCoalSpecs\_Rev4\_20130510.pdf (accessed in 2015).
- Park, S.S., Kozawa, K., Fruin, S., Mara, S., Hsu, Y.-K., Jakober, C., Winer, A., Herner, J., 2011. Emission factors for high-emitting vehicles based on on-road measurements of individual vehicle exhaust with a mobile measurement platform. J. Air Waste Manag, Assoc. 61, 1046—1056.
- PSCAA (Puget Sound Clean Air Agency), 2005. 2005 National Air Toxics Assessment (accessed in 2015). http://www.pscleanair.org/library/Documents/3-10-11% 202005%20NATA%20info%20sheet.pdf.
- Ramanathan, V., Carmichael, G., 2008. Global and regional climate changes due to black carbon. Nat. Geosci. 1, 221–227.
- Seinfeld, J.H., 1986. Atmospheric Chemistry and Physics of Air Pollution. Wiley, New York
- Sierra Research, 2004. Revised Inventory Guidance for Locomotive Emissions. No. SR2004-06-01. http://www.metro4-sesarm.org/pubs/railroad/FinalGuidance.pdf (accessed in 2015).
- U.S. DHHS (U.S. Department of Health and Human Services), 2014. 13th Report on Carcinogens (accessed in 2015). http://ntp.niehs.nih.gov/pubhealth/roc/roc13/ index.html.
- U.S. EPA (U.S. Environmental Protection Agency), 2014. List of Designated Reference and Equivalent Methods (accessed in 2015). http://www.epa.gov/ttnamti1/files/ ambient/criteria/reference-equivalent-methods-list.pdf.
- U.S. EPA (U.S. Environmental Protection Agency), 2013. Locomotives (accessed in 2015). http://www.epa.gov/otaq/locomotives.htm.
- U.S. EPA (U.S. Environmental Protection Agency), 2012. Report to Congress on Black Carbon. EPA-450/R-12-001. (accessed in 2015). http://www.epa.gov/blackcarbon/2012report/fullreport.pdf.
- U.S. EPA (U.S. Environmental Protection Agency), 2009. Emission Factors for Locomotives. EPA-420-F-09-025. (accessed in 2015). http://www.epa.gov/nonroad/locomotv/420f09025.pdf.
- von der Weiden, S.L., Drewnick, F., Borrmann, S., 2009. Particle loss calculator a new software tool for the assessment of the performance of aerosol inlet systems. Atmos. Meas. Tech. 2, 479–494.
- WA DOE (Washington Department of Energy), 2013. Environmental Review: Gateway Pacific Terminal at Cherry Point Proposal (accessed in 2015). http://www.ecy.wa.gov/geographic/gatewaypacific.
- Wang, X.L., Chancellor, G., Evenstad, J., Farnsworth, J.E., Hase, A., Oslon, G.M., Sreenath, A., Agarwal, J.K., 2009. A novel optical instrument for estimating size segregated aerosol mass concentration in real time. Aerosol Sci. Technol. 43, 939—950.
- Wang, Y.G., Hopke, P.K., Rattigan, O.V., Xia, X.Y., Chalupa, D.C., Utell, M.J., 2011. Characterization of residential wood combustion particles using the two-wavelength aethalometer. Environ. Sci. Technol. 45, 7387–7393.
- Watson, J.G., Chow, J.C., Lowenthal, D.H., Pritchett, L.C., Frazier, C.A., Neuroth, G.R., Robbins, R., 1994. Differences in the carbon composition of source profiles for diesel-powered and gasoline-powered vehicles. Atmos. Environ. 28, 2003–2005.
- WHO (World Health Organization), 2012. IARC: Diesel Engine Exhaust Carcinogenic. Press Release No. 213. (accessed in 2015). http://www.iarc.fr/en/media-centre/pr/2012/pdfs/pr213\_E.pdf.
- Yanowitz, J., McCormick, R.L., Graboski, M.S., 2000. In-use emissions from heavyduty diesel vehicles. Environ. Sci. Technol. 34, 729–740.
- Zhu, J.P., Aikawa, B., Pigeon, R., 2005. Measurement of fine particles in diesel emissions using a real-time aerosol monitor. J. Air Waste Manag. Assoc. 55, 978–983.