Anonymous Referee #1
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We appreciate the reviewer's insightful comments. They have clearly improved the manuscript. For reviewer's information, we have added new Supplementary Information (SI) to the end of our responses here.

1. General

1.1   The authors develop a curve fit for 'dispersion coefficients' using a Gaussian dispersion model solution. However there is no real motivation given for using this approach rather than using dispersion models directly (e.g. ADMS-Roads, AERMOD, Caline etc.).

➢ Thank you for pointing out this potential source of confusion. The purpose of this study is not to evaluate/improve "the performance of existing near-road dispersion models"; rather it is to investigate how routinely measured variables affect plume magnitude, transport, and concentration decay rates, and consequently, to evaluate the areal impact of traffic plumes from major roadways. This would be very difficult to do with a sophisticated model, for the reasons outlined below. To make quantitative comparison between measured explanatory variables and plume characteristics (including plume peak concentrations and concentration decay shape downwind), it is effective to define and extract quantitative (numerical) plume parameters that explain well the plume characteristics observed from measured concentration profiles. This is the reason we applied a curve fit method using Gaussian dispersion model solutions rather than using dispersion models.

Current sophisticated dispersion models require a comprehensive input dataset including, for example, friction velocity, surface heat flux, boundary layer and mechanical mixing heights (particularly during daytime under more turbulent conditions) surface roughness, vertical wind profiles, complex roadway configurations, etc. These requirements prevent direct and quantitative comparison between input variables and observed concentration profile shapes, regardless of the model performance. Moreover, detailed turbulence measurements are not readily and routinely conducted in urban areas, and unfortunately were not collected in this study. Although many modeling studies estimate turbulence

parameters based on Monin-Obhukov similarity theory, the theory is not perfect for the real atmosphere.

Finally, our study focuses on plume behavior in the stable nocturnal boundary layer, which is typically the most challenging situation for current dispersion models. We also note that for the sampling periods (4:30 – 6:30 A.M.), traffic flows in the target highways are sharply increasing due to the morning commute, which is, we think, difficult to represent even with the current comprehensive models.

➤ To make this clearer, in the revised manuscript we added a short section to explain the reasons why we used a curve fit method and the advantages (recognizing there are limitations) of this method over sophisticated dispersion modeling studies (in Section 1 Introduction and Section 2.3.1 Development of an optimized formulation). Now it reads as below:

In the Introduction,

"Many studies have attempted to predict the pollutant concentrations from vehicular emissions near roadways using different dispersion models with varying levels of complexity (Sharma and Khare, 2001). However, most studies have focused on predicting elevated pollutant concentrations at specific distances from sources rather than describing concentration profiles. A few studies attempted to reproduce UFP concentration profiles obtained at multiple discrete distances within short ranges (<300 m) during daytime conditions (Zhu and Hinds, 2005;Gramotnev et al., 2003;Heist et al., 2013).

Gaussian dispersion models have been commonly used to explain spatial concentration variations from line sources (e.g., Sharma and Khare, 2001;Chen et al., 2009;Briant et al., 2011;Gramotnev et al., 2003;Kumar et al., 2011;Heist et al., 2013). Simple Gaussian models require parameterization of dispersion coefficients, which is critical to calculate pollutant concentrations at specific distances from the source. Existing parameterizations of the dispersion coefficients for these models are based on Pasquill stability classes (Pasquill, 1961). However, the Pasquill parameterization has only two classes for stable conditions (Table 1), and thus has limited ability to explain the variations in concentration profiles under stable conditions. In addition, current sophisticated dispersion models (such as AERMOD, CALINE, RLINE etc.) require a comprehensive dataset including friction velocity, surface heat flux, boundary layer and mechanical mixing heights, surface roughness, vertical wind profiles, complex roadway configurations, etc. (Heist et al., 2013). Detailed turbulence measurements as well as boundary layer and mechanical mixing heights and surface roughness, etc. cannot be readily and routinely obtained in urban areas. In addition, these requirements prevent direct and quantitative comparison between input variables and observed concentration profile shapes, regardless of the model performance. Although many modeling studies estimate turbulence parameters based on Monin-Obhukov similarity theory, the theory is not perfect for the real atmosphere and particularly under stable nocturnal conditions, which is typically the most challenging situation for current dispersion models (Heist et al., 2013). We also note that for our sampling periods (4:30 to 6:30), traffic flows in the target highways are sharply increasing due to the

morning commute, which is, we think, difficult to represent even with the current comprehensive models. While these more complex sophisticated models are ideal for many applications, here we probe how routinely measured variables (basic meteorological parameters and concentration data) affect UFP plume shapes, and thus have chosen to use a modified Gaussian expression for our investigation.

In the present study, the objectives are to investigate how routinely measured variables affect UFP plume magnitude, transport, and concentration decay rates and consequently, to evaluate the areal impact of traffic plumes from major roadways. For this reason, the effectiveness of the Gaussian dispersion model solution to fit observed UFP concentration profiles is examined, and both dispersion coefficients and emission factors are obtained directly from the observations in this study. In addition, the quantitative effects of surface meteorological parameters and the role of concentration differences between plumes and backgrounds on plume extensions are investigated. Appropriate parameterization of dispersion coefficients and emission factors based on observable variables can provide predictive capability for the extent of freeway plumes under stable conditions."

And in section 2.3.1; esp. see additional text at the end:

"A Gaussian dispersion model solution assuming an infinite line source was applied as a basic expression, and as fits to the observed concentration profiles (Eq. 1):

$$C(x,z) = \frac{Q}{\sqrt{2\pi}\sigma_z(x) \cdot U_e} \left[\exp\left(-\frac{(z+H)^2}{2\sigma_z^2(x)} \right) + \exp\left(-\frac{(z-H)^2}{2\sigma_z^2(x)} \right) \right] \tag{1}$$

where Q is an emission rate, U_e is an effective wind speed (ambient wind + speed correction due to traffic wake), z is height, H is the height of the emission source, and σ_z is the standard deviation of the time-averaged concentration distributions in the vertical direction at distance x from the source (Luhar and Patil, 1989). An infinite line source assumption is reasonable for the present study due to the long length of freeways (more than 20 km) compared to relatively short downwind length scale of transects (~2 km). A simple Gaussian dispersion model solution was chosen as a basic equation to minimize the number of free variables to fit to the observations, leading to results that are consistent and reliable and can be effectively interpreted.

Equation (1) is simplified to obtain a final modified Gaussian expression (Eq. 2), where Q_c represents a bulk emission parameter including emission rate (Q) combined with wind effects (U_e), and remains as a free variable to be determined from observed concentration profiles.

$$C(x,z) = \frac{Q_c}{\sigma_z} \left[\exp\left(-\frac{(z+H)^2}{2\sigma_z^2} \right) + \exp\left(-\frac{(z-H)^2}{2\sigma_z^2} \right) \right] \tag{2}$$

The final step to formulate a simplified model equation is to parameterize σ_z. For this, two common methods were examined: Chock's (1978) and Briggs' (1973) formulas, which were used by Luhar and Patil (1989) and Briant et al. (2011), respectively, in their model evaluations. However, we note that both Chock's and Briggs' formulas have just one or two equations for stable atmospheres, based on land use (e.g., urban and rural). Thus, neither formula is sufficient to explain the meteorology-dependent variations in observed freeway plume decay during stable pre-sunrise hours. To account for these limits, two coefficients in Chock's and Briggs' formulas were held as free variables in the modified Gaussian equation (e.g., α and β for Briggs formula in Eq. 3). We found that the Briggs' formula form more successfully described the observed concentration profiles. The Briggs expression has slightly different formulations for rural and urban conditions (Table 1), the choice of which affects one of the two dispersion coefficients (β). Both forms fit the equally data well and produce nearly identical curve shapes. For three of our four transects the dispersion coefficients returned by the formulation β is more consistent with the rural form (described more below). While this may seem surprising, much of Los Angeles,

including these three transects, consist of single story residential development. The fourth transect, DTLA, has tall buildings in the area (although few tall buildings are on the transect itself), and its β values are closer to expected urban values. Here, we use the rural form of the Briggs' formula as the basic equation for fitting the observations, to allow us to investigate meteorological and traffic effects on plume intensities and transport among the different sites. More discussion of the observed α and β are presented in *section 3.1*.

$$\sigma_z(x) = \frac{\alpha \cdot x}{1 + \beta \cdot x} \tag{3}$$

Fitted results using Chock's formula tended to underestimate the peak concentrations near freeways. Additionally, we examined a K-theory model developed by Sharan and Yadav (1998) for dispersion of pollutants from a point source under stable conditions with light winds (Table 1). Zhu and Hinds (2005) modified the K-theory model for a line source to explain the decay of a freeway plume during daytime. The K-theory model yielded poorer fits to our nocturnal observations in the far downwind areas compared to the Gaussian model with an optimized Briggs formulation for σ_z above. Consequently, Eq. (2) combined with Eq. (3) was used to fit the observed data using the least squares method. This formulation, a modified Gaussian dispersion expression with an optimized Briggs formulation (optimized by fitting observational data) is hereinafter referred to as the "*modified GB model*."

We acknowledge that this modified GB model does not explicitly consider the traffic related turbulence or the surface roughness effects on dispersion. However, vehicle-induced turbulence is relatively short-lived, and has a dominant effect in the immediate vicinity of the roadways (Wang and Zhang, 2009;Gordon et al., 2012), becoming negligible within 60 m downwind from the roadways (Gordon et al., 2012). This range covers only a small fraction of our UFP profile range (up to 2 km). In addition, vehicle-induced turbulence likely varies little between our sampling sites and over our measurement time periods, for two reasons; first because trucks and passenger cars induce markedly different turbulence, significant differences in vehicle fleets could result in differences in turbulence. In our study however diesel trucks consistently contributed less than 6% of the total traffic for all freeways. Second, for the pre-sunrise periods, vehicle speed among all sampling days and sites due to the consistent free-flow of traffic. As described in SI.1, all sites investigated had similar built-environments (i.e., transects were surrounded mostly with 1-story residential single-family houses). Thus, we believe the surface roughness should be similar among our sampling sites."

1.2   Indeed there is no actual discussion/consideration of these models at all which does seem very odd. Given this, it is not clear what the study has achieved nor what insight it has given into 'factors controlling plume length (as the title implies)' – there needs to be much clearer statement of this. Overall the paper lacks coherence and purpose and needs to be much improved.

➢ While we certainly agree such models can be valuable tools in many studies, we respectfully disagree that using current sophisticated models is the only option for studies of near-roadway pollutant distributions and transport. We adopted Gaussian model solutions to extract important but simple plume shape parameters at the ground level directly from our observed profiles, and there is no further dispersion modeling work in the manuscript. Our simple statistical modeling is solely based on direct comparisons between extracted plume parameters from observed profiles and observed meteorological

and traffic variables. As a result it does not seem appropriate to discuss/consider these sophisticated models in the manuscript: this would seem to be more appropriate for a separate study/manuscript. We have addressed this point as well in a new section (see also response to 1.1 above) describing the value of our approach and its contrast with more sophisticated models.

1.3   There seems to be no conclusions/discussion. This should be added and at least cover the general applicability of the dispersion coefficients or otherwise. Can the work be applied elsewhere?

➢   We appreciate the reviewer pointing this out and encouraging us to expand on this important point. We added additional discussion in the revised manuscript in the last Section in response to this point. Our results showed consistent relationships between observed plume parameters and simultaneously observed meteorological and traffic variables for stable pre-sunrise periods at four different sites in the South Coast Air Basin of California. These results imply that we can apply our results to other urban/suburban residential areas under stable pre-sunrise conditions with calm but consistent wind patterns. We have addressed this point in the last paragraph of Section 3.6 and now it reads:

In section 3.6,

"Nonetheless, we consequently believe this approach provides an efficient and precise tool to predict freeway plume profiles near major roadways under stable conditions in that: (1) dispersion parameters can be extracted directly from the real atmosphere; (2) these simple dispersion parameters explain the observed UFP concentration profiles, producing excellent agreement for all sampling sites; (3) quantitative and straightforward comparisons between plume parameters and controlling meteorological/traffic factors can be made; (4) the considered conditions (the onset of morning commute with increasing traffic in stable air) are difficult to represent even with the current comprehensive models; (5) multivariate regression results can be applied with readily and routinely measurable variables without sophisticated model expertise. Although investigated environments were limited in this study (nocturnal calm stable conditions in residential areas) and hence one cannot expect that our results can be applied directly to other environments with different surface roughness and air stability, our results have potential implications given that many residential areas near freeways/highways have similar built environments (at least in the U.S.) and nocturnal stable conditions are common. Particularly we note that about 50% of the population lives within 1.5 km of freeways in the South Coast Air Basin of California (Polidori et al., 2009). This study also provides useful datasets and the potential to parameterize dispersion coefficients and emission factors for more sophisticated model simulations."

1.4   Contrary to the 'General Guidelines for Manuscripts & Submission' some sections of the paper do not have good sentence structure. For example, the paragraph below equation (4) in Section 3.4 has one sentence beginning 'Because: : :', and the following sentence begins 'In addition,

because: : :'. There are also many instances of 'we' and 'our'; this reviewer would prefer these sentences to be re-worded to use a passive tense.

➤ We are flexible with the writing style and regret that the reviewer found our style choice problematic. We note that we have not been able to find guidelines prohibiting sentences from beginning with 'because' or using 'we'/'our'. Further, use of the active voice is encouraged by most journals today. Nonetheless, we are willing to additionally revise the manuscript to avoid the active voice if the editor prefers, although the expressions the reviewer pointed out above have been corrected accordingly.

1.5   There are many instances where a comparative adjective has been used to describe a noun, and it is not clear what the noun is being compared against, for example, the first sentence in section 3.4 'Hypothesis 2 states that more intensive plumes can decay faster due to larger concentration gradients between background and plume.' – more than what? Finally, a number of sentences begin with a formula (for example, the beginning of Section 3.4.1). This reviewer would like some effort to be made to re-word these sentences so that they begin with a word, and the formula is introduced later in the sentence.

➤ We revised the manuscript accordingly. We adopt such concise constructions partly in response to the pressure to meet word limits. ACP doesn't have word limits, so perhaps the style is not appropriate in this case.

2. *Specific comments/questions:*

2.1   2.1 Section 1.0 – The Share and Khare 2001 is an old reference, it would be helpful to reference more recent work (refer to, for instance, the special editions of the International Journal of Environment and Pollution which are published alongside the Harmo meetings http://www.harmo.org/, and the recent work by Heist et al. in Transportation Research Part D: Transport and Environment 'Estimating near-road pollutant dispersion: A model inter-comparison').

➤ This suggested reference has been added in the revised manuscript.

2.2   Section 2.1 – It would be useful to add map of area where data was collected with transects marked.

➤ An appropriate map was added in the Supplementary Information (SI. 1)

2.3   Section 2.2 – On p25258, line 8, a distance is given in non - SI units.

➤ All non-SI units were corrected in the revised manuscript, including the emission factor.

2.4   Section 2.3.1, Equation (1) – It would be good to comment on why a term taking into account reflections of the plume at the top of the boundary layer has been omitted from this expression. This term may be important in conditions where there are night time surface inversions.

➢ The concept of a boundary layer top is one that applies during common daytime conditions; however it is not relevant to discussion of nocturnal surface inversions, which are the focus of this manuscript. A daytime convective boundary layer is commonly capped with a stable layer above, and this stable layer can reflect a plume back toward the ground. In contrast, for our cases, the nocturnal boundary layer is stably stratified from the surface to several tens of meters. The thickness of this layer typically grows as the night progresses. Above the stably stratified surface layer is a neutral layer referred to as the residual layer, in which turbulence produced during the previous daytime persists. Pollutants emitted from the surface are strongly inhibited from mixing vertically within the nocturnal boundary layer, and as a result are not expected to be well mixed. This temperature structure creates the large, persistent plumes studied here. If pollutants were to reach the top of the nocturnal surface layer, they would be expected to be easily incorporated and dispersed into the neutral residual layer above, not reflected back toward the surface.

➢ We leave it to the editor to decide if he feels this needs to be explicitly explained in the manuscript, depending on his judgment concerning the background of the expected readership.

2.5   Section 3.1 – The first sentence says that the curve fits provide excellent matches to the observations. Are we really surprised by this, given that the curves have been derived from the observed data?

➢ It is not clear to us what the reviewer's concern is here. The statement related to the match of the fits to the shape of the decay curves is important to the discussion that follows, but we agree it doesn't rise to the level of 'surprising'. It isn't obvious either however, as detailed below. The implications of the excellent agreement of the curve fits and observations are as follows. First, the simple Gaussian model solution form with Briggs' formula serves well, and allows the subsequent extraction of the plume parameters as discussed in the manuscript. As mentioned in the manuscript, we also tried other forms (Chock's formula and the K-theory model) for the dispersion coefficient σ_z and found the Briggs' form performed better than the others. Second, the Gaussian model

was fit to high spatial resolution (~ 20 m) continuous concentration profiles, a more stringent test than previous daytime studies made mostly with only three to five discrete points. Thus the result is somewhat novel.

Third, excellent matches mean that we can describe the plume shape/length including peak and far downwind concentrations well enough if we know just three plume parameters (Q_c, α and β). This can make straightforward and quantitative the complicated links between dispersion/transport of highway pollutant plumes and meteorological, traffic, and geographical conditions.

Fourth, the excellent agreement with observations supports the notion that if Q_c, α and β extracted from the observations are reasonable, it is possible to make quantitative and straightforward comparisons between these parameters and related explanatory variables such as meteorological and traffic variables.

Consequently, if we can obtain these three plume parameters properly with a simple statistical tool (in this study, the multivariate regression method was used), we can predict reasonably well the plume magnitude and concentration drop-off rates as well as downwind concentrations with readily and easily measurable variables and without the use of comprehensive and sophisticated dispersion models that should be operated by sophisticated experts.

2.6   Section 3.2 – What is plume intensity?

➢ Plume intensity means both the magnitude of pollutant concentrations denoted as $\Delta[UFP]$ and plume peak width. Thank you for pointing this out; we clarified the term plume intensity in the revised manuscript (in the first sentence of Section 3.4 and 3.4.1) as below:

"Hypothesis 2 states that plume decay rates (here likely dilution rate) are a function of concentration gradients between plumes and backgrounds: as the concentration differences between plumes and backgrounds become larger, concentration decreases faster with time."

And

"The plume intensity parameter $\Delta[UFP]_{peak}$, defined as the differences between the background and plume peak concentrations, showed clear and consistent negative correlations with both the dispersion coefficients α and β (Fig 6a and b), in contrast to wind speed and direction."

2.7   Section 3.3 - This is a rather poor description of factors controlling dispersion which are well known. The descriptions given do not give confidence that the authors really understand these processes; they should be improved.

> We agree these are incomplete descriptions of factors controlling dispersion. In this section we attempted to provide a short conceptual description for Hypothesis I (winds effects) to explain the strong positive relationship we observed between α and β, before discussing/showing results in the following sections (*Sect. 3.3.1* and *3.3.2*), however we will work to improve the descriptions in this section to address the concerns of the reviewer. We acknowledge that the title of the *Sect. 3.3* is broad, and thus changed the title to narrow the scope of our discussions to: "Wind effects on plume characteristics". It now reads:

"Hypothesis 1 states that pollutants can be effectively advected farther with relatively moderate winds blowing steadily in one direction under stable conditions. Stronger winds in the surface layer may produce more turbulence due to stronger wind shear and also deepen the mechanical mixing length to disperse pollutants more rapidly through effective eddy-diffusion processes. Thus, for stable pre-sunrise hours, moderate and consistent winds may be able to effectively transport plumes (smaller α), but would result in faster decay rates (smaller β), compared to weaker winds. Thus, wind effects on extracted plume parameters are examined in this section."

<div align="center">**Supplementary Information**</div>

<div align="center"># Factors controlling pollutant plume length downwind of major roadways in nocturnal surface inversions</div>

<div align="center">W. Choi, A. M. Winer, and S. E. Paulson</div>

SI.1 Characteristics of sampling areas (Downtown LA, Paramount, Carson, and Claremont)

To investigate the areal impact of freeway plumes on nearby residential neighborhoods under stable pre-sunrise conditions, four different measurement sites were selected in the South Coast Air Basin (SoCAB) in California: Downtown Los Angeles (DTLA), Paramount, Carson and Claremont (Fig. S1a). The SoCAB occupies a coastal plain surrounded by mountains on three sides (the San Gabriel, San Bernardino, and San Jacinto mountains). The predominant meteorological conditions in the SoCAB are characterized by mild winds and shallow boundary layer heights capped by low-altitude (500 to 1200 m above ground level) temperature inversions due to a semi-permanent "Pacific High" pressure cell. Prevailing winds are dominated by diurnal cycles of weak off-shore breezes at night and stronger on-shore sea breezes during the day. Nighttime surface cooling combined with weak winds often builds up a stable layer at the surface and up through the first ~200 m of the lowest edge of the atmosphere. This shallow nocturnal surface layer prevents air ventilation and hence accumulating vehicular emissions.

The four sampling routes ("transects") were about 3 to 4 km long (1 to 2 km upwind and 2 to 2.5 km downwind of the freeways). Each aligned as close to perpendicular as possible to straight sections of freeway. The freeways were roughly perpendicular to prevailing winds and away from interchanges with other freeways or major arterials. Each transect ran along quiet, residential two-lane streets surrounded (as much as possible) with one-story single houses (Fig. S1b). None of the chosen transects had direct freeway access; this greatly reduces interference from local high-emitting vehicles and traffic in general. Sampling transects passed: *under* the 101 freeway in Downtown Los Angeles (DTLA), *under* the 91 freeway in Paramount, *over* the I-110 freeway in Carson, and *over* the I-210 in Claremont (Fig. S2).
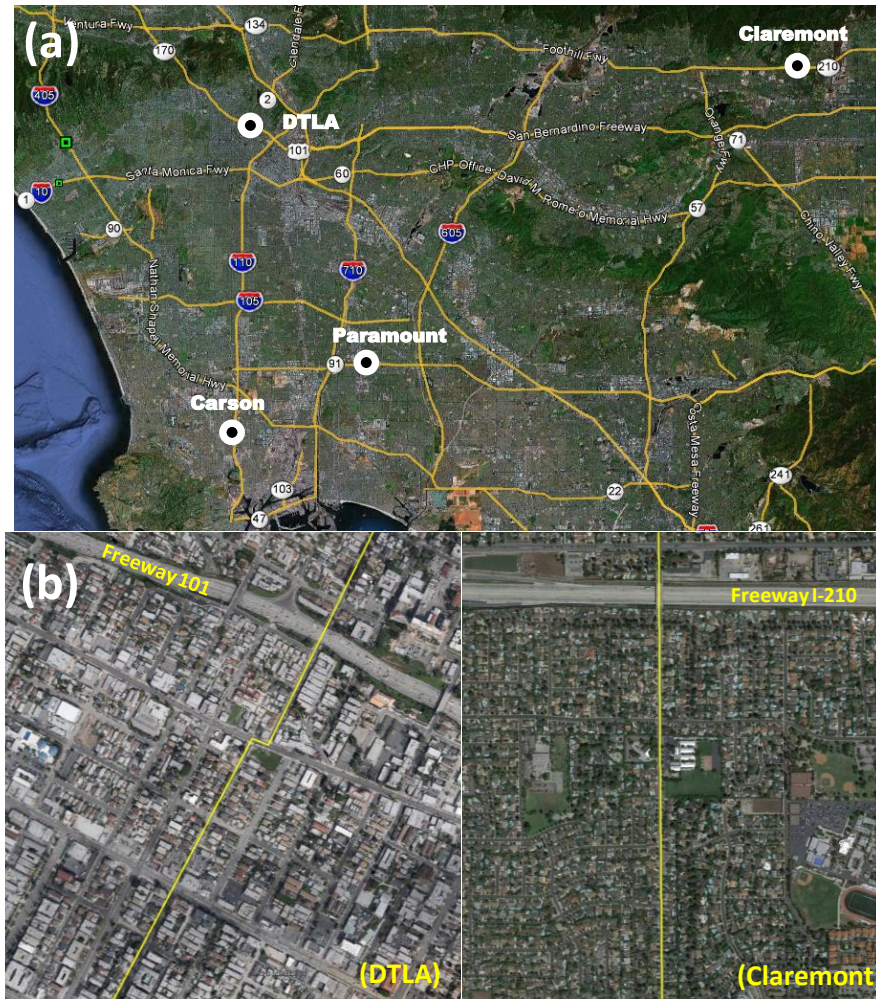
Fig. S1. (a) Map of transect locations where pre-sunrise measurements were conducted in the South California Air Basin (SoCAB). (b) Close up maps of transects the mobile platform drove on (yellow lines) and surroundings around the transects in DTLA (bottom left) and Claremont (bottom right). *Google Earth* map.
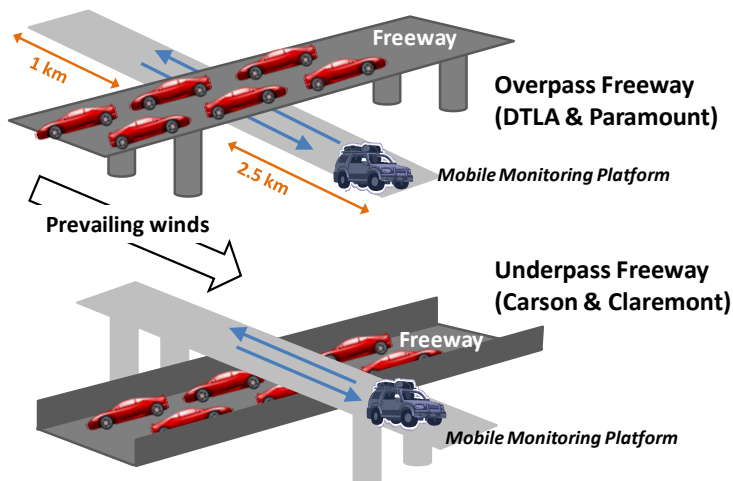

Fig. S2. Schematic illustration for freeway-transect geography for overpass (top) and underpass freeways (bottom). Sketch does not represent the scale of geographical features.

The DTLA transect, near downtown Los Angeles, follows N. Coronado St., a small two lane street, running north–south. The entire upwind area and first 1500m of the downwind area is residential. The farthest 1500 to 2200 m on the downwind side traverses a commercial district with tall buildings. The Paramount transect is located 11 km from the coast in a part of the coastal plain and is surrounded entirely by residential areas. The Carson transect is also on the coastal plain, ~ 6 km northwest of the Ports of Los Angeles and Long Beach. The transect is mostly surrounded by residential areas, however the upwind end of the Carson (> 850 m from the freeway) and downwind ends of Paramount (> 1400 m from the freeway) are adjacent to industrial/commercial areas. We did not find evidence of pollutant emissions from these industrial areas in our measurements as might be expected particularly in the pre-sunrise hours. Finally, the Claremont transect is located in an inland valley, ~70 km from the coast at the foot of steeply rising San Gabriel Mountains. The transect is entirely surrounded by quiet residential areas. The DTLA transect is crossed by several arterial streets downwind of the freeway: Temple St., Beverly Blvd., 3rd St., 6th St., and Wilshire Blvd. The Carson, Paramount and Claremont transects each are crossed by just one or two major streets: Figueroa St. and Main St. for the Carson transect, Artesia Blvd. for the Paramount transect, and Foothill Blvd. for the Claremont transect. However, only small numbers of vehicles were observed on the cross streets during the pre-sunrise measurement periods. Nonetheless, to avoid possible interference from local vehicular emissions on these cross streets, data obtained in the vicinity (several tens meters on the downwind side) of these streets were excluded from our analyses. Some parts of above descriptions were taken from Choi et al. (2012;2013).

SI.2. Instrumentations, sampling, and post-data processing

A Toyota RAV4 electric sub-SUV was used as mobile monitoring platform (MMP) to avoid self pollution. The MMP was equipped with a suite of fast response instruments for various air pollutants: CPC 3007 and FMPS for ultrafine particle number concentrations; DustTraks for $PM_{2.5}$ and PM_{10}; PAS 2000 for particle-bound polycyclic aromatic hydrocarbon; gas pollutants monitoring including CO, NO and CO_2. The MMP was also equipped with a GPS (Garmin 76CS) for MMP position and a 2D-sonic anemometer for winds; and temperature and humidity sensors (Choi et al., 2012). Spatial distributions for other pollutants near the freeways were described in

more detail in Choi et al. (2012). The same MMP has been used in a number of studies conducted in the SoCAB and the detailed instrumentation and calibration information is available elsewhere (Hu et al., 2009;Kozawa et al., 2012;Westerdahl et al., 2005;Choi et al., 2012). Briefly, air was pulled through a 6" diameter galvanized steel manifold installed through windows of the rear passenger space (1.5 m a.g.l.) by a fan located downstream of all sampling ports. Sampling ports for each instrument are located in the middle of manifold with short (0.5 to 2m) sampling tubing (1/4" Teflon for gases and 1/4" conductive tubing for particles and 1/2" conductive tubing for FMPS). Particle and gas instruments were calibrated by their respective manufacturers just before field measurements began. Calibration checks for gas-instruments were also conducted before each sampling campaign. Flow and zero checks were conducted on a daily basis. Data were recorded using a data-logger (Eurotherm Chessell Graphic DAQ Recorder) with 1 second time resolution, which corresponds to 5 to 8 m spatial resolution when the MMP was driving at 20 to 30 km/h normal driving condition during measurements.

The MMP was driven along transects during pre-sunrise periods (4:30 to 6:30), for most sampling days the last run was completed just before sunrise time, making about 6 profiles (scans) per day in general. The sparse local traffic on the transect allowed the MMP to be driven at the same low speed (less than 30 km/h) through the whole transect, so that fine spatial resolution of concentration profiles could be obtained (5 to 8 m). Once sampling was completed, cross-correlation method (Eq. S1, Choi et al., 2012) was applied on a daily basis to correct the different response time of each instrument in the MMP, which was caused by the characteristics of the instruments themselves and the length and flow rates through their inlets. Several smoke tests were also conducted as a reference.

$$r = \frac{1}{T\sigma_a\sigma_b} \int_{t_0}^{t_0+T} \left(a(t) - \bar{a}\right)\left(b(t+\tau) - \bar{b}\right)dt \qquad \text{(Eq. S1)}$$

where a and b are simultaneously measured species, t is time, τ is a time-lag applied to time series in b, σ is the standard deviation for the two pollutants a and b, and T is the number of data points in the time-series. Data synchronization using cross-correlation worked effectively given that traffic-related pollutants are emitted concomitantly from vehicles and reach peak concentrations near the sources, e.g., major roadways (Choi et al., 2012). After synchronizing instrument response time, local transient spikes in spatial concentration profiles from nearby

high-emitting vehicles were removed by a running low 25% quantile method with varying window sizes (Choi et al., 2012): 53 s for distance farther than 1 km from the freeway; 31 s for distance between 300 m and 1 km; and 3 s within 300 m from the freeways. This method successfully removed transient local spikes without altering remaining data. We additionally examined any remaining local effects, particularly near freeways, by reviewing video and audio records to verify proximity of high emitting vehicles before removing corresponding data.

A balloon tether sonde (SmartTether[TM], Anasphere Inc.) was used to probe the vertical temperature, humidity and wind gradients to determine atmospheric stability. Vertical profiles (up to ~ 100 m a.g.l.) for temperature, humidity and winds were obtained on a daily basis (about 30 minutes before the MP measurements) near the transects (560 m away from the Downtown LA transect, 1.2 km from the Paramount transect, 3.7 km from the Carson transect, and 3.8 km from the Claremont transect). It was not possible to launch the balloon immediately adjacent to the transects due to air safety regulations (balloon launches are prohibited within 8 km of any airport) as well as the requirement for adequate open space to launch a balloon.

SI.3. General Meteorology and Traffic Conditions for Measurement Periods

The usual prevailing wind direction was approximately perpendicular to the freeway for the DTLA, Paramount, and Carson transects with mean directions in the 73 to 82° range relative to the freeways (90° being normal to the freeway orientation). For the Claremont transect, winds were more askew to the freeway with a mean direction of 58°. Winds for this transect were the least variable however, due to the adjacent mountains to the north which produce a strong, thermally-induced, mountain-valley wind system. Wind speeds during the sampling periods were generally calm (0.3 to 1.1 $m \cdot s^{-1}$ for all sampling days). Investigated areas were influenced by weak off-shore breezes at night and stronger daytime on-shore wind shift in general occurs around 9 A.M. in the summer and later in the winter. Thus, the measurement sites have experienced consistent winds for pre-sunrise measurement periods. Temperature varied day-by-day, ranging from 3 to 15 °C, but varied little (within ±0.5 °C) during our short early morning sampling period. Static atmospheric stability can be represented with a vertical potential temperature gradient ($d\Theta/dz > 0$ for stable, $d\Theta/dz \sim 0$ for neutral, and $d\Theta/dz < 0$ for unstable).

During the measurement periods, dΘ/dz was slightly positive for all transects indicating slightly stable conditions. The vertical temperature gradient was highest near the Claremont transect (1.23 $\times 10^{-2}$ K·m^{-1}) although the differences by location were not significant. Winds were generally calm with little vertical gradient during the measurements periods although the Claremont transect showed relatively stronger wind gradient compared to the other sites making the air more neutral in terms of Richardson number (Fig. 8b).

The MMP measurements were conducted during the period of sharply increasing traffic flow on the freeways due to the onset of the morning commute. The 5 minute traffic and truck flows on the freeways were obtained from the Freeway Performance Measurement System (PeMS) sensors in the vicinity of the sampling transects: 100 m northeast of the DTLA transect (VDS ID: 717452); 550 m east of the Paramount transect (VDS ID: 765467); 850 m south of the Carson transect (VDS ID:763522); and 60 m east of the Claremont transect (VDS ID:767984). The mean traffic flows during the measurement periods were 800, 1000, 630, and 470 vehicles per 5 minutes on the 101 (DTLA), 91 (Paramount), I-110 (Carson), and I-210 freeways (Claremont), respectively. The fleet mixes on the transects were not characterized in detail; however they were not obviously different from one another. Truck flows accounted for a small fraction of the total traffic flows, falling in a similar range for all transects (2.4 to 6%, Table 3 in Choi et al. (2012)). The differences in truck contribution should result in moderate differences in mixed-fleet emission rates for each transect, as well as between our measurements and those in the literature.

References

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