Anonymous Referee #2
Received and published: 29 December 2013

We have carefully considered the referee's concerns and revised the manuscript accordingly. At the same time, we leave the line-by-line responses to referee's comments in this document.

1. This is an interesting article that uses some of the data collected during earlier work (Choi et al 2012) of the authors and the UFP data is assessed here. While the article provides some interesting curve-fitting results, there are a number of technical concerns as well as transparency issues in writing and presenting the article. For instance, (i) it is hardly mentioned where measurements were taken – where these measurements were mobile or stationary at fixed locations? (ii) How does the measurement locations suitable for applying the standard Gaussian equations. These details of measurements are important but missing and referred to another paper – some top level information should be included. A schematic depicting the site and the measurements location would assist readers to understand these clearly.
   - First, we agree that some top level information on sampling sites and methods are not included in the manuscript and the reader is referred to another paper (Choi et al., 2012). (i) In Choi et al. (2012) details about sampling methods and four measurement sites as well as data post-processing algorithms, including response-time-synchronizing between instruments and spatial resolution of UFP profiles, are described. We thought initially this repeated information (published in Choi et al., 2012) was too lengthy to be placed in this “part II” article where much more extensive data analyses have been conducted. However, we agree with the referee's concerns and have added a brief version of this information to the text and have added more details in the Supplementary Information (SI. 1 through 3). The SI information is attached to the end of this response.
   - (ii) Our conclusion that measurement locations are suitable for applying the standard Gaussian equations was led by the curve fit results. If the Gaussian equation does not work as a basic equation for fitting observed concentration profiles, reproduced profiles would not show a good agreement with observations; in this case clearly the Gaussian equation works very well. Please see "Responses 2.5 to Referee #1" for more discussions about curve fit results. Detailed discussions were added in the first paragraph of both Section 2.3.1 and Section 3.1.

2. Authors are using the data in parts in various papers and as a result a lot of important information which authors seems to keep for a follow up article (Choi et al. 2013) is missing and crucial for the

development of the arguments. The authors should first clearly present the measurements and their analysis before moving to their modeling and data fitting.

2.1. We apologize for this confusion. Choi et al., 2012, already published, contains the requested information. Key parts of the material have also been added to the Supplementary Information section associated with this manuscript.

Moreover, as authors mentioned by themselves, this is a too simple approach to deal with the UFP modelling – using a simple Gaussian equation without taking into account the (i) transformation processes which are likely to affect the concentrations notably because of high UFP concentrations, and (ii) any treatment of the traffic related turbulence as well as the roughness effects is too simplistic, raising serious concerns about the practical applicability of the results obtained.

2.2. (i) Although particle dynamics can affect particularly particle number concentrations, there is universal agreement that dilution/dispersion plays the most important role in UFP concentration decreases with distance accounting for about 90% of the total loss of UFP number (Kumar et al., 2011) under convective daytime conditions. In an earlier study of the same area (West Los Angeles) Jacobson and Seinfeld (AE, 2004) suggested that 2% of the decrease in particle concentration was due to coagulation and most of the remaining 98% was due to dilution during daytime.. In our case, dilution plays a somewhat smaller role as the timescales for plume dispersion are longer in the stable early morning. As mentioned in the manuscript, β values likely represent additional UFP number losses besides dilution/dispersion, particularly including particle dynamics. This is another advantage of our analytical approach, not a disadvantage; current dispersion models have been developed initially based on non-reactive conservative species, and hence particle dynamics are not modeled perfectly (Zhang et al., 2004; Jacobson and Seinfeld, 2004; Jacobson et al., 2005). In fact, many sophisticated models targeting UFP show less satisfactory results particularly under stable conditions (Heist et al., 2013). Although our analysis treats particle dynamics effects implicitly in this study, these additional effects are represented in β values of the "real" atmosphere (which were extracted directly from the observations). We have attempted to explain how these dispersion parameters are related to concurrently measured meteorological parameters. Discussions about particle transformation issue have been addressed in the revised manuscript (in the last paragraph of Section 2.3.1 and Section 3.1). The last paragraph of section 2.3.1 now reads as follows:

"Although particle number concentrations are influenced by particle dynamics such as coagulation, deposition, and condensation/evaporation, a common conclusion from previous studies is that dilution is the most important process controlling particle number (e.g., Kumar et al., 2011;Zhang et al., 2004;Jacobson and Seinfeld, 2004). Particularly near emission sources, such as the curbside of a major road, the dilution timescale is approximately one to two orders of magnitude faster than deposition and coagulation, respectively (Kumar et al., 2011). Even under stable nocturnal conditions, dilution appears to be the most important sink accounting for ~70% of the overall decay rates (Choi and Paulson, in prep). Nonetheless, we note that dispersion parameter β (as discussed in *3.1*) represents additional UFP number losses besides dilution/dispersion due to these transformation processes because here dispersion parameters (α and β) were extracted by fitting the observed UFP profiles of the "real" atmosphere. We believe this is another advantage of our analytical approach because current dispersion models have been developed initially based on non-reactive conservative species, and hence particle dynamics are not perfectly modeled in the real atmosphere (Zhang et al., 2004;Jacobson and Seinfeld, 2004)."

And a short addition to section 3.1:

"and (4) although it is implicit, additional loss processes due to particle dynamics are considered in β, which to our knowledge is not considered perfectly in current dispersion models applied to the real atmosphere. In any case, the model fitting methods provide an effective tool to estimate dispersion coefficients directly from the observations."

(ii) Vehicle-induced turbulence lives for a short time and has a dominant effect in the immediate vicinity of the roadways (Wang and Zhang, 2009; Gordon et al., 2012). Both Wang and Zhang (2009) and Gordon et al. (2012) showed traffic-induced turbulence became negligible within 300 m downwind from the roadways. Particularly, based on observations, Gordon et al. (2012) reported that turbulence kinetic energy elevated from the roads had almost disappeared within 60 m downwind. This covers a very small part of our UFP profile range (up to 2 km). We acknowledge that the observed peak concentrations could be affected somehow by vehicle-induced turbulence. However, we do not think vehicle-induced turbulence varies much at our sampling sites and over our measurement time periods because (1) turbulence produced by passenger cars is much smaller than that from trucks, and for all freeways studied here diesel trucks consistently contributed < 5% of the total traffic; and (2) for the pre-sunrise periods, vehicle speeds are consistent due to free-flow of traffic. Thus we think vehicle-induced turbulence did not vary significantly between freeways studied here. In addition, we defined a particle number emission factor as "effective particle number that survived (or “net concentration”) the initial mixing and particle dynamics processes", because our current knowledge cannot separately quantify the effects of nucleation, coagulation, condensation/evaporation, as well as initial mixing processes, on particle number emissions at the initial "tail-pipe to roadway" scale.

With regard to surface roughness, all measurement sites investigated in this study had similar built-environments (i.e., freeways are surrounded mostly with 1-story residential single houses) because one of our study objectives is to investigate the areal impact of freeway plumes over residential areas. Thus, we believe the surface roughness should be similar at the various sampling sites. However, we appreciate the referee pointing out that one cannot expect that our results can be applied directly to open grass fields, forests, or possibly centers of cities with very tall buildings. Nonetheless we think most residential areas near freeways/highways (at least in the U.S.) likely have similar built environments and our results can be reasonably applied to these residential areas. We have added more details about the measurement sites as described above. These issues have been addressed in the second to last paragraph of Section 2.3.1 and the last paragraph of Section 3.6:

From Section 2.3.1:

"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."

And 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."

3. The authors treat the UFP as a single entity based on their numbers but the dispersion behaviour of nucleation (<30 nm) and accumulation mode particles is completely different and should not be dealt as same.
    - ➢ Unfortunately the term "accumulation mode" is used in different ways in the literature, so we aren't certain we have understood the reviewer's point correctly. However the most conventional definition of accumulation mode particles is 100 – 300 nm. These particles contribute minimally to the total UFP number concentrations because vehicular emissions are concentrated mostly in the nucleation mode (10 – 30 nm).
    - ➢ With regard to dispersion, all of the small particles of interest here should behave very similarly. The smallest particles have somewhat higher Brownian diffusion rates, however for our situation (time scales of ~ 1 hr) diffusion has a negligible effect on dispersion of all particles; the Brownian diffusion length scale is much smaller than those associated with diffusion caused by atmospheric eddies. Eddy diffusion should not act differently on nucleation and larger particles because eddy diffusivity is determined by atmospheric turbulence states (Stull, 1998; Seinfeld and Pandis, 1998). The more rapid diffusion rates of nucleation mode particles do facilitate removal processes via enhanced coagulation and/or dry deposition. However, (1) the dispersion parameters of our analyses implicitly represent these particle dynamics effects combined with dispersion/dilution processes, and (2) these particle dynamics processes are not explicitly revealed even in sophisticated model (please see also above).

4. Effect of temperature is discussed, though this does not seem to be mentioned anywhere in the article, what was the temperature and its variation. Given that the measurements are considered during the morning hours and therefore a huge temperature difference is likely to occur during measurements. How did author apportion this effect?
    - ➢ We appreciate the reviewer pointing out the need to be more explicit about temperature. Discussions concerning temperature effects in this manuscript are based on Fig. 7 (b),

although no values were presented in the text or tables. We have added the mean and the range of observed temperatures during measurement periods to Table 2.

If the sampling periods had covered the entire morning, we agree that large temperature differences would be expected. However, because the measurements were conducted for one and half to two hours before sunrise (4:30 to 6:30 A.M.), the temperature each day was nearly constant, varying within only ±0.5 °C. More discussions about general meteorological characteristics for measurement periods have been added in Supplementary Information SI. 3.

5. Given the way the data used is presented and standard approach applied to UFP modelling and considering the usefulness of the results obtained, the article need much more in-depth thinking and analysis to make it useful for the readers.

    - More specifics (such as which sort of analysis or things to consider) would have been helpful with this comment. Nonetheless, we respectfully disagree; we believe the article can contribute significantly to understandings of (a) the areal impact of ultrafine particles emitted from the freeways during stable pre-sunrise periods; (b) the characteristics of how fast UFP concentrations decrease; (c) how these decay rates, as well as the peak concentrations, are linked with meteorological/traffic conditions for pre-sunrise periods; and (d) how we can effectively predict these UFP plume impacts on residential areas under common stable pre-sunrise conditions.

Supplementary Information

Factors controlling pollutant plume length downwind of major roadways in nocturnal surface inversions

W. Choi, A. M. Winer, and S. E. Paulson

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).

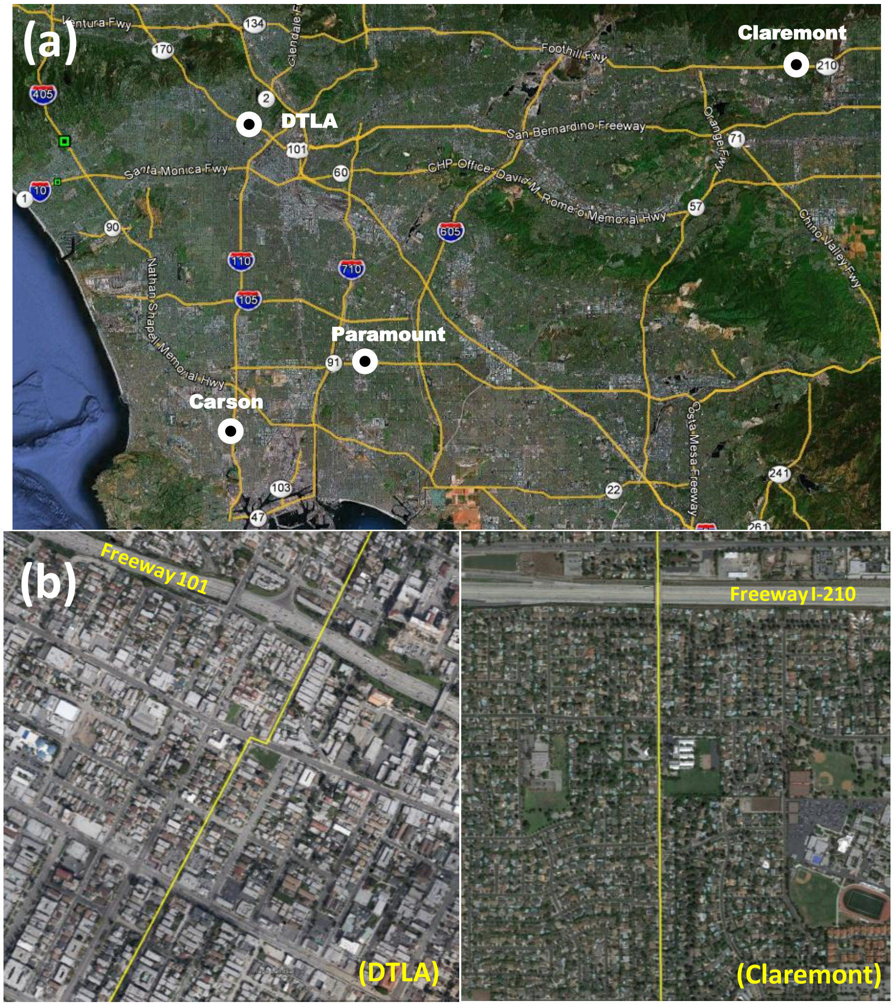

Fig. S1. (a) Map of transect locations where pre-sunrise measurements were conducted in the South California Air Basin (SoCAB). (b) Close up maps of transects the mobile platform drove on (yellow lines) and surroundings around the transects in DTLA (bottom left) and Claremont (bottom right). *Google Earth* map.

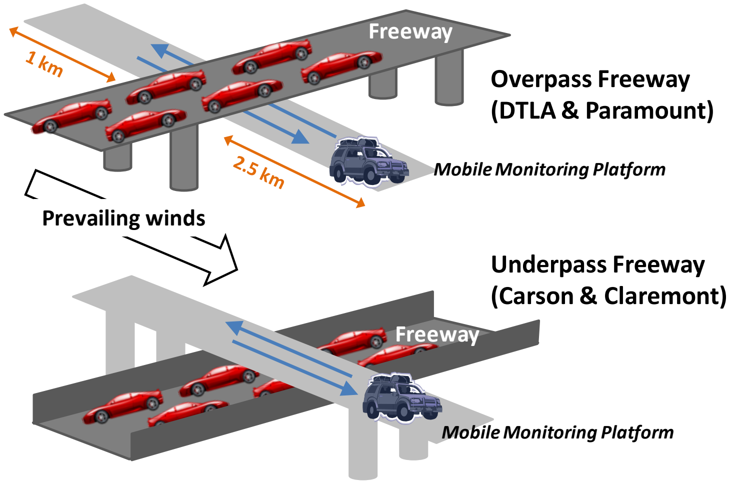

Fig. S2. Schematic illustration for freeway-transect geography for overpass (top) and underpass freeways (bottom). Sketch does not represent the scale of geographical features.

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 andCO_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™, 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 (1.23×10^{-2} $K \cdot m^{-1}$) 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

Choi, W., He, M., Barbesant, V., Kozawa, K. H., Mara, S., Winer, A. M., and Paulson, S. E.: Prevalence of wide area impacts downwind freeways under pre-sunrise stable atmospheric conditions, Atmospheric Environment, 62, 318-327, http://dx.doi.org/10.1016/j.atmosenv.2012.07.084, 2012.

Choi, W., Paulson, S. E., Casmassi, J., and Winer, A. M.: Evaluating meteorological comparability in air quality studies: classification and regression trees for primary pollutants in California's South Coast Air Basin, Atmospheric Environment, 64, 150-159, http://dx.doi.org/10.1016/j.atmosenv.2012.09.049, 2013.

Hu, S. S., Fruin, S., Kozawa, K., Mara, S., Paulson, S. E., and Winer, A. M.: A wide area of air pollutant impact downwind of a freeway during pre-sunrise hours, Atmospheric Environment, 43, 2541-2549, 10.1016/j.atmosenv.2009.02.033, 2009.

Kozawa, K. H., Winer, A. M., and Fruin, S. A.: Ultrafine particle size distributions near freeways: Effects of differing wind directions on exposure, Atmospheric Environment, 63, 250-260, 2012.

Westerdahl, D., Fruin, S., Sax, T., Fine, P. M., and Sioutas, C.: Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles, Atmospheric Environment, 39, 3597-3610, 10.1016/j.atmosenv.2005.02.034, 2005.