Observation of Ions and Particles near Busy Roads using a Neutral Cluster and Air Ion Spectrometer (NAIS)

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Abstract

Motor vehicles emit large quantities of ions in the form of both charged particles and molecular cluster ions. While, the health effects of inhalation of charged particles is largely unexplored, the concentrations near busy roads and the distance to which these particles and ions are carried have important implications for the exposure of the large percentage of the population that lives close to such roadways. We measured ion concentrations using a neutral cluster and air ion spectrometer (NAIS) near seven busy roads carrying on the average approximately 7000 vehicles hr\(^{-1}\) including about 15% heavy duty diesel vehicles. In this study, charged particle concentrations were measured as a function of downwind distance from the road for the first time. We show that, at a moderate wind speed of 2.0 m s\(^{-1}\), mean charged particle concentrations at the kerb were of the order of 2x10\(^4\) cm\(^{-3}\) and, more importantly, decreased as \(d^{0.6}\) where \(d\) is the distance from the road. While cluster ions were rapidly depleted by attachment to particles and were not carried to more than about 20 m from the road, elevated concentrations of charged particle were detected up to at least 400 m from the road. Most of the charge on the downwind side was carried on the larger particles, with no excess charge on particles smaller than about 10 nm. At 30 nm, particles carried more than double the charge they would normally carry in equilibrium. There are very few measurements of ions near road traffic and this is the first study of the spatial dispersion of charged particles from a road.

Keywords: Ion, charged particle, vehicle emission, pollution, dispersion
1. Introduction

Atmospheric ions may be classified by size and type into two broad categories. These are ‘small ions’ or ‘cluster ions’ which are charged molecular clusters smaller than 1.6 nm and ‘large ions’ which are essentially charged particles larger than this size. Under stable conditions, cluster ions are continually formed by the ionization of air molecules by cosmic radiation and natural radioactivity and lost by recombination and attachment to aerosols. They naturally occur in concentrations of about 100-200 cm$^{-3}$ (Reiter, 1992) but may be up to an order of magnitude greater in the presence of anthropogenic ion sources such as overhead high voltage power transmission lines (Buckley et al., 2008; Jayaratne et al., 2008) and motor vehicle traffic (Jayaratne et al., 2010; Titta et al., 2007). Typical cluster ion concentration in the urban background is about 300 cm$^{-3}$ (Ling et al., 2010; Retalis et al., 2009). Under stable conditions with no ion sources, approximately 1 in 3 aerosol particles carry a charge, the vast majority being a single charge. In areas away from ion sources, this will result in concentrations of 500-1000 cm$^{-3}$. However, in the presence of an ion source, this may increase to several thousand cm$^{-3}$, with many particles carrying more than one charge (Jayaratne et al., 2011; Lee et al., 2012).

Motor vehicles are known to emit both cluster ions and charged particles of both signs with total concentrations in the exhaust gases being of the order of 10$^6$ - 10$^7$ ions cm$^{-3}$ for petrol engines and 10$^8$ ions cm$^{-3}$ for diesel engines (Collings et al., 1988; Yu et al., 2004). About 80% of the particles in diesel emissions are charged, with roughly equal amounts of positive and negative charge with the number of charges per particle
increasing with particle size from one charge at 40 nm to four charges at 300 nm (Kittleson et al., 1986; Maricq, 2006). Thus, we expect there to be high concentrations of both cluster ions and charged particles near busy roads.

(Jayaratne et al., 2010) showed that the mean total cluster ion concentration near city roads (600 cm\(^{-3}\)) was about one-half of that near freeways (1200 cm\(^{-3}\)) and about twice as high as that in the urban background (270 cm\(^{-3}\)). Both positive and negative cluster ion concentrations near a freeway showed a significant linear increase with traffic density and correlated well with each other in real time. Cluster ion concentrations decreased sharply with downwind distance immediately near the freeways, the rate of decrease being significantly greater than for particles. At one site investigated, within the first 10 m from the road, the cluster ion concentration decreased by more than 70\%, while the particle number concentration decreased by just 10\%. In general, measured cluster ion concentrations decreased to background within about 15 m from the kerb. This was expected because cluster ions emitted by motor vehicles have a very limited lifetime, quickly attaching to particles present in the exhaust. Therefore, near busy roads, most of the emitted charge would be expected to reside on particles.

(Titta et al., 2007) used an air ion spectrometer to monitor cluster ions and charged nanoparticles about 10 m away from a busy freeway carrying 14,000-16,000 vehicles per day in Finland. They found average cluster ion concentrations of 320 cm\(^{-3}\) and 280 cm\(^{-3}\) for negative and positive ions respectively. The corresponding charged nanoparticle concentrations were 750 cm\(^{-3}\) and 510 cm\(^{-3}\), respectively. They compared these values with measurements obtained around the same time at a rural, clean air
station and showed that the cluster ion concentration at this site was about three-times higher than that at the traffic site, while the nanoparticle charge concentration was only about one-third as much.

(Lee et al., 2012) used a tandem differential mobility analyser to monitor particles of four different mobility diameters (30, 50, 80 and 100 nm) carrying up to 3 charges each, as a function of distance from two busy freeways. They measured the fraction of particles carrying a given charge and showed that the fractional decay of the charged particles with distance increased with the number of charges carried. They did not monitor cluster ion concentrations nor did they specify the concentrations of the charged particles in the environment.

Motor vehicles are the main source of air pollution in urban environments and the emitted particles are accompanied by large concentrations of ions. Owing to the strong interaction between these two, it is not easy to predict the balance between cluster ions and charged particles and how they vary with distance from a busy road. The presence of ions and charged particles in the air is of concern as it has been suggested that they may be linked to several adverse health effects such as respiratory and cardiological conditions (Fews et al., 1999; Henshaw, 2002). Therefore, it is important to investigate the nature and concentration of these charges near roads and to determine how far they would be carried from a road and for how long they would last in the environment. With the rapidly increasing urban sprawl and the close proximity of residential areas to busy roads the findings in this study have an important bearing on human exposure to ions.
In this study, we used a neutral cluster and air ion spectrometer (NAIS) to measure positive and negative cluster ions and charged nanoparticle concentrations simultaneously as a function of distance from busy roads for the very first time and show that they follow very different trends.

2. Methods

2.1 Instrumentation

The neutral cluster and air ion spectrometer (NAIS) was developed by Airel Ltd, Estonia. It is designed to monitor neutral and charged aerosol particles and clusters in the mobility range 3.16 to 0.001 cm² V⁻¹ s⁻¹, which corresponds to a particle size range of 0.8 to 42 nm. A detailed description of its operation may be found in (Manninen et al., 2009; Mirme et al., 2007). Controlled charging and electrostatic filtering enables the NAIS to differentiate neutral aerosols from charged ions. It uses two parallel spectrometer columns, one of each polarity, so that both positive and negative ions may be monitored simultaneously. As the air is drawn into the instrument, the particles are charged and passed into the two columns where they are classified by a cylindrical differential mobility analyzer. Ions are deflected in a radial field and collected on 21 electrically isolated electrometer rings according to their electrical mobilities. The ion currents from the rings are measured with electrometers, providing ion and particle number distributions in selected size fractions. The instrument switches between ion and neutral particle modes by cycling the operation of the chargers and electrostatic filters in user-defined time periods. In this study, we set the NAIS to operate in a cycle of 3.0 min including ion and neutral particle sampling periods of 2.0 and 0.5 min, respectively, the remaining 0.5 min being an
offset period which is required to neutralize and relax the electrodes. The total sampling air flow was 60 L min\(^{-1}\), the high flow rate being used to minimize ion diffusion losses and maximize the measured ion concentration sensitivity. Ion losses are accounted for during post-processing of the data by the software (Mirme et al., 2007). Readings were logged at 1 s intervals.

The upper size detection limit of the NAIS effectively restricts it to the nanoparticle size range. Particles in vehicle emissions, particularly soot, have a mode between 50 and 100 nm. Larger particles can carry a greater charge than smaller particles so that a substantial portion of the charges reside on particles larger than 42 nm that are not detected by the NAIS. Therefore, the particle charges reported in this study may be considered to be lower limits and not the total charges carried by the particles present.

2.2 Field Measurements

Measurements were carried out at seven different sites near busy freeways around Brisbane, Australia (Locations and details of the sites are given in Fig S1 and Table S1). The traffic density on the freeways was 130-160 vehicles min\(^{-1}\) with 10-20% being heavy-duty diesel vehicles. The NAIS was placed on a laboratory trolley, 2-5 m away from the edge of the road, with its sampling inlet at a height of 0.8 m above the ground. At one of the sites, with the wind blowing normal to the freeway, measurements were repeated at varying distances in the downwind direction. The selected site comprised of level ground with short grass cover, well away from trees and other obstacles. The trolley was moved along a line normal to the road, with measurements obtained at regular intervals along this line, up to a maximum distance
of 400 m. Data were logged for periods of 10-15 min at each location. Measurements were also obtained on the upwind side of each freeway immediately before and after the experiments and these were treated as the respective background values.

The NAIS was powered by a portable petrol-powered ac voltage generator which was placed at least 10 m away in the downwind direction from the instruments. Wind speed was estimated using a hand-held anemometer, while the wind direction was noted on a fixed weather vane. Traffic density was estimated manually by counting the number of light and heavy-duty vehicles passing in each 1 min period at intervals of five minutes. Air temperature and humidity were recorded at regular intervals over the measurement periods.

2.3 Data analysis

All data were obtained at 1 s intervals. In deriving representative values of the parameters at a given location, both mean and median values were considered. The mean values were generally skewed because both the particle number and charge concentrations monitored near the freeways showed large spikes associated with emission plumes from heavy duty diesel vehicles. When comparing magnitudes between different locations, mean values were used so that the data points could be treated statistically using the Student’s t-test. Linear regression analyses were used to investigate significant increases or decreases of parameters. All significant differences in both the t-tests and the regression analyses were estimated at a confidence level of 95%.
3. Results and Discussion

3.1 Kerbside Measurements

Fig 1 shows a typical short time series of the total charge particle and total cluster ion concentrations at one of the seven measurement sites located 2 m away from the kerb of a freeway. The mean traffic density during the measurement period was 152 min\(^{-1}\), including 20 min\(^{-1}\) heavy duty vehicles. The measurements were conducted on the downwind side of the freeway. The wind velocity varied between 3 and 9 km h\(^{-1}\) and the angle between the road and the wind was between 45\(^\circ\) and 90\(^\circ\).

This figure gives an indication of the relative magnitudes of the charges carried by clusters and particles near a busy road. The general trends and magnitudes of charge observed at all seven kerbside locations were similar. The gaps between the clustered data points in the figure are due to the cyclic operation of the NAIS as described in the methods section. Note the very high variability of the ion concentrations in time, which was typical at close proximity to motor vehicles. At this site, the mean total particle charge concentration over a period of 90 min was 1.95 \(\times\) 10\(^4\) cm\(^{-3}\) with a standard deviation of 1.01 \(\times\) 10\(^4\) cm\(^{-3}\). The corresponding median value was 1.72 \(\times\) 10\(^4\) cm\(^{-3}\). At all seven sites, the mean total particle charge concentration was 1.7 \(\times\) 10\(^4\) cm\(^{-3}\) with a standard deviation of 9.5 \(\times\) 10\(^3\) cm\(^{-3}\). Large spikes in concentration often exceeded 5.0 \(\times\) 10\(^4\) cm\(^{-3}\) and coincided with the passage of large trucks, especially with the wind blowing from the road. Cluster ion concentrations also showed large spikes that generally coincided with the charged particle spikes. The corresponding mean and median total cluster ion concentrations at this site were 595 and 406 cm\(^{-3}\), respectively. The mean value from all seven sites was 640 cm\(^{-3}\), which indicates that
the mean charge carried by the particles was at least 25 times larger than that carried by cluster ions.

Both the charged particle and cluster ion concentrations on the downwind side showed large rapid fluctuations due to vehicular traffic emissions while there were no such variations on the upwind side (Fig S1). An interesting observation was that, while the charged particle concentration on the downwind side was always greater than on the upwind side, the cluster ion concentration on the downwind side was often less than that on the upwind side. We attribute this to the attachment of cluster ions to particles that were present in high concentrations on the downwind side.

### 3.2 Polarity of charges

Next, we look at the relationship between the positive and negative ions. On the upwind side, a paired Student’s t test showed that the mean positive cluster ion concentration was significantly higher than the negative value. However, the two quantities were not positively correlated ($R^2=0.01$, $p>0.05$). Fig S3 shows a plot of the positive charged particle concentration against the negative value over a short time period. Each point is a 5 s data point. Again, a paired t test showed that the mean positive charge concentration was significantly higher than the negative value but they did not show a positive correlation to each other ($R^2=0.26$, $p>0.05$).

From the time series plots of the ion concentrations observed on the downwind side, it was immediately obvious that there was a very strong correlation between the two polarities for the charged particles with the two time series curves exhibiting a near-
perfect mirror image. Positive and negative spikes generally coincided in time, suggesting that they were from the same source. This is consistent with the findings of (Kittleson et al., 1986) and (Maricq, 2006) that motor vehicle exhaust contains approximately equal numbers of positively and negatively charged particles. In a previous study (Jayaratne et al., 2010), we reported a similar relationship between positive and negative cluster ions near roadways. The correlation between positive and negative cluster ion concentrations is not as strong as it is for charged particles.

These observations may be explained in terms of the relative mobility of the two types of ions. Negative clusters have a higher mobility than positive clusters and, therefore, a higher efficiency of ion neutralization and attachment (Kolarž et al., 2009). Thus, their lifetime and concentration are generally lower than positive ions in stable environments such as on the upwind side of the road (Tammet et al., 2006). However, at the downwind monitoring sites, the ions and charged particles emitted by vehicles do not have a sufficiently long time to attain equilibrium and, since ions of the two polarities are produced in equal numbers, the concentrations of positive and negative ions, whether they be clusters or particles, show a strong correlation to each other.

In Fig 2, we present a plot of the downwind positive charged particle concentration against the corresponding negative value over a short time period. Each point is a 5 s data point. In contrast to the upwind plot, shown in Fig 3, the downwind plot shows a strong correlation between the positive and negative charge concentrations, with $R^2=0.9$ and $p<0.05$. Further, unlike in the upwind case, a paired t test showed that there was no significant difference between the mean values of the two quantities.
3.3 Ion size distribution

Next, we look at the size distributions of the ions. Fig 3 shows the median concentrations of negative and positive ions measured at the downwind site classified into nine size bins as shown. The smallest bin (< 1.6 nm) corresponds to cluster ions. The median negative and positive cluster ion concentrations were 142 and 153 cm$^{-3}$, respectively. Similar to what was observed for cluster ions, in most of the size bins, the median positive and negative charged particle concentrations were not significantly different. The median negative and positive charged particle concentrations were $8.7 \times 10^3$ cm$^{-3}$ and $8.9 \times 10^3$ cm$^{-3}$, respectively. Fig S4 presents the same information for the upwind location. Here, the median negative and positive cluster ion concentrations were 240 and 281 cm$^{-3}$, respectively. The median negative and positive charged particle concentrations were $1.35 \times 10^3$ cm$^{-3}$ and $1.57 \times 10^3$ cm$^{-3}$, respectively.

In Fig 4, we compare the median size distributions of the total ion concentration on the upwind and downwind sides of the freeway. The overall total ion concentrations on the upwind and downwind sides were $2.90 \times 10^3$ cm$^{-3}$ and $1.76 \times 10^4$ cm$^{-3}$, respectively. It is clear that the fluctuation of ion concentration is greater on the downwind side. On the upwind side, the peak ion concentration occurred in the 5-10 nm size bin, while on the downwind side the particles were larger, with a peak size in the 25-30 nm size bin. The corresponding total particle number size distributions are shown in Fig S5. Fig 5 shows the ratio of the ion concentrations on the downwind and upwind sites in the nine size bins. It is clear that most of the charges are on the larger particles. This is consistent with the relative ion-aerosol attachment coefficients at the
different sizes (Buckley et al., 2008). For example, for ions with mobility of 1.2 cm² V⁻¹ s⁻¹ and mass 150 amu, the coefficient for a neutral 10 nm particle is 0.220 x 10⁻⁶ cm³ s⁻¹, while for a neutral 50 nm particle it is 1.87 x 10⁻⁶ cm³ s⁻¹ (Hoppel and Frick, 1986). Therefore, cluster ions are more likely to attach to the larger particles in the dispersing exhaust plume of a vehicle.

Aerosol particles are charged by naturally occurring cluster ions and attain an equilibrium charge distribution that follows an approximate Boltzmann function. The larger the particle, the greater the charge that it can hold and, thus, the fraction of aerosols that are charged increase with size, from less than 10% at 10 nm to about 57% at 100 nm (Fuchs, 1963; Wiedensohler, 1988). However, near an ion or particle source, such as a busy road, the aerosols are not in charge equilibrium. Therefore, it is useful to express their charge in terms of the equilibrium charge of aerosols of the same size. In Fig 6, the curve shows the percentage of particles that are charged in equilibrium as reported by (Fuchs, 1963; Wiedensohler, 1988). The open triangles show the experimental values determined by the NAIS from the ratio of the number of charged particles to the number of total particles (charged plus neutral) at each size bin. The closed circles show the overcharging ratio, defined as the ratio of the measured percentage of particles charged to the percentage of particles charged in equilibrium. Thus, an overcharging ratio of 1.0 indicates that the particles do not carry any excess charge over what they would be expected to carry in charge equilibrium. It is apparent that, near the freeway, particles smaller than about 10 nm do not carry any excess charge, while the overcharging ratio increases with particle size beyond 10 nm, reaching a value greater than 2.0 at 30 nm.
3.4 Variation with distance

Next, we look at the variation of ion concentrations with downwind distance from the freeway. Fig 7 shows the mean positive and negative cluster ion concentrations up to a distance of 400 m from the freeway. At the closest approach of 14 m from the freeway, the positive and negative cluster ion concentrations were 380 and 210 cm$^{-3}$. From 14 m to 25 m, the concentrations dropped dramatically to 100 and 90 cm$^{-3}$, respectively. This is due to the rapid attachment to particles in this region and is in good agreement with Jayaratne et al (2010), who showed that the cluster ion concentrations dropped rapidly within the first 20 m from the freeway. Thereafter, as the PNC decreased with distance, the cluster ion concentration gradually recovered, reaching about 250 and 150 cm$^{-3}$ at 400 m. While the positive cluster ion concentration has regained its upwind background value (240 cm$^{-3}$), the negative value is still about one-half of the background value (280 cm$^{-3}$).

Fig 8 shows the charged particle concentrations as a function of distance from the freeway. The variations of the mean positive and negative charged particle concentrations were remarkably similar, showing power law decreases of $d^{-0.61}$ ($R^2=0.98$) and $d^{-0.64}$ ($R^2=0.98$), respectively, where $d$ is the distance from the freeway. At 400 m from the freeway, the charged particle concentrations, although near background, were still decreasing. We conclude that elevated particle charge concentrations extended up to about 400 m from the freeway.

These results were obtained at a steady wind speed of 2.0 ± 0.5 m s$^{-1}$. The distances to which charged particles are carried depend on the wind speed and at higher speeds it...
is likely that they are carried further afield (Jayaratne et al., 2010). It is instructive to observe that the total charged particle concentration found near the kerb ($1.9 \times 10^4$ cm$^{-3}$) was more than an order of magnitude higher than that found in the only other study of charged particle concentrations near a traffic road ($1.3 \times 10^3$ cm$^{-3}$) (Titta et al., 2007). However, we note that the traffic density in the present study was about 15 times greater than that in the Titta et al study. Moreover, the density of heavy-duty vehicles in our study was about 20 min$^{-1}$, whereas there was minimal heavy-duty in the Titta et al study.

4. Conclusions

We conclude that most of the charges on the downwind sides of busy roads are carried on the larger particles, with no excess charge on particles smaller than about 10 nm. At 30 nm, particles carried more than double the charge they would normally carry in equilibrium. The charged particle concentration close to the kerb was at least 25 times greater than the cluster ion concentration. The total charged particle concentration on the downwind side of the freeway was about six times higher than on the upwind side. The mean charged particle concentrations of both signs decreased with distance, $d$, from the freeway, following a power law $d^{-0.6}$. Elevated particle charge concentrations extended up to about 400 m from the freeway. Cluster ions readily attach to particles so that their concentration dropped rapidly within the first 25 m from the freeway. Thereafter, as the particle concentration decreased, it gradually recovered, reaching its background value at about 400 m.

With the urban spread in many cities, the residential population living and working in close proximity to busy freeways is rapidly increasing. Studies have shown that
smaller particles can penetrate deep into the human lungs while larger particles are mostly trapped in the upper airways of our respiratory system, such that the probability of deposition of inhaled particles in the alveolar region of the lung is a maximum in the particle size range 10-20 nm (Hinds, 1999). Studies have also shown that charged particles have a greater probability of deposition in the lungs than uncharged particles of the same size (Cohen and Xiong, 1998) so that there is a considerable risk of exposure to charged particles in the size range measurable by the NAIS (< 42 nm). Therefore, the results of this study, in particular the concentrations of ions and charged particles near busy roads and the distances to which they are carried, are important in assessing the potential risks associated with the inhalation of motor vehicle emissions.

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References


Figure Captions

Fig 1: Total charged particle and cluster ion concentrations near a busy freeway.

Fig 2: Downwind 5 s positive charged particle concentrations against the corresponding negative values.

Fig 3: Median size distributions of negative and positive ions measured at the downwind site. The error bars represent the Q3 and Q1 percentiles.

Fig 4: Median size distributions of the total ion concentrations measured at the upwind and downwind sites. The error bars represent the Q3 and Q1 percentiles.

Fig 5: The downwind/upwind ratio of ion concentrations as a function of particle size.

Fig 6: Percentage of particles charged (Δ) and the overcharging ratio (○) as a function of particle size as determined from the equilibrium particle charge distribution (shown by the line graph).

Fig 7: Mean positive and negative cluster ion concentrations as a function of distance from the freeway.

Fig 8: Mean positive and negative charged particle concentrations as a function of distance from the freeway.
Supplementary Data

The Supplementary Data accompanying this paper includes five figures and one table. One paragraph of descriptive text accompanies Fig S2.

The captions to the five figures are listed below.

Supplementary Data Figure Captions

Fig S1: Map showing the locations of the seven monitoring sites (from Map data ©2013 GBRMPA, Google). See Table S1 for details.

Fig S2: Short time series of (a) charged particle and (b) cluster ion concentrations measured upwind and downwind at the kerb of a busy freeway.

Fig S3: The relationship between the positive and negative charged particle concentrations on the upwind side of the road. The straight line shows equality.

Fig S4: Size distributions of negative and positive ions measured at the upwind site. The error bars represent the Q3 and Q1 percentiles.

Fig S5: Median size distributions of the total particle number concentrations measured at the upwind and downwind sites. The error bars represent the Q3 and Q1 percentiles.
Fig S1
**Table S1**: Details of the seven measurement sites as indicated in Fig S1.

Measurements as a function of distance from the road were conducted at site 3.

<table>
<thead>
<tr>
<th>Site</th>
<th>Motorway</th>
<th>Suburb</th>
<th>#Lanes</th>
<th>Latitude (S)</th>
<th>Longitude (E)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Pacific Mwy</td>
<td>Loganholme</td>
<td>6</td>
<td>27º 40’ 53.8”</td>
<td>153º 11’ 12.8”</td>
</tr>
<tr>
<td>2</td>
<td>Pacific Mwy</td>
<td>Springwood</td>
<td>6</td>
<td>27º 37’ 47.3”</td>
<td>153º 07’ 51.4”</td>
</tr>
<tr>
<td>3</td>
<td>Gateway Mwy</td>
<td>Tingalpa</td>
<td>4</td>
<td>27º 28’ 44.6”</td>
<td>153º 07’ 00.4”</td>
</tr>
<tr>
<td>4</td>
<td>Bruce Hwy</td>
<td>Burpengary</td>
<td>4</td>
<td>27º 10’ 25.0”</td>
<td>152º 58’ 54.5”</td>
</tr>
<tr>
<td>5</td>
<td>Bruce Hwy</td>
<td>Burpengary</td>
<td>4</td>
<td>27º 09’ 24.0”</td>
<td>152º 58’ 32.5”</td>
</tr>
<tr>
<td>6</td>
<td>Bruce Hwy</td>
<td>Morayfield</td>
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<td>152º 58’ 38.6”</td>
</tr>
<tr>
<td>7</td>
<td>Bruce Hwy</td>
<td>Caboolture</td>
<td>4</td>
<td>27º 02’ 39.8”</td>
<td>152º 58’ 38.2”</td>
</tr>
</tbody>
</table>
Fig S2
Text to accompany Fig S2:

Fig S2 compares the charged particle and cluster ion concentrations on the downwind side of the freeway with that on the upwind side. The upwind and downwind measurements could not be conducted at the same time as there was only one NAIS available. Therefore, the upwind measurements were obtained soon after the downwind measurements, ensuring that the mean traffic density and wind direction were not significantly different. The data gaps, resulting from the sampling cycle of the NAIS as described in the Methods section, have been closed up. Thus, the upwind and downwind data in Fig S2 do not match up in time. However, the charged particle data in Fig S2(a) are synchronous with the cluster ion data in Fig S2(b).
Fig S3
Fig S4

Fig S4 (in Black and White)
Fig 1

Particle Size Bin (nm)

Median Particle Number Conc (cm$^{-3}$)

- Upwind
- Downwind

Fig 2

Concentration (ions cm$^{-3}$)

Time

Charged Particles

Cluster Ions
Fig 3 Colour

Fig 4
Fig 5

Downwind/Upwind Ratio vs. Particle Size Bin (nm)

Fig 6
Fig 7

Fig 8
$y = 53,607.07x^{-0.64}$
$R^2 = 0.98$

$y = 48,010.50x^{-0.61}$
$R^2 = 0.98$