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Knibbs, Luke D., Cole-Hunter, Tom, & Morawska, Lidia (2011) A review of commuter exposure to ultrafine particles and its health effects. *Atmospheric Environment*, 45(16), pp. 2611-2622.

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<http://dx.doi.org/10.1016/j.atmosenv.2011.02.065>

1 Review paper

2

3 **A review of commuter exposure to ultrafine particles and its health effects**

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26 **ABSTRACT**

27 Ultrafine particles (UFPs, <100 nm) are produced in large quantities by vehicular combustion  
28 and are implicated in causing several adverse human health effects. Recent work has  
29 suggested that a large proportion of daily UFP exposure may occur during commuting.  
30 However, the determinants, variability and transport mode-dependence of such exposure are  
31 not well-understood. The aim of this review was to address these knowledge gaps by  
32 distilling the results of 'in-transit' UFP exposure studies performed to-date, including studies  
33 of health effects.

34

35 We identified 47 exposure studies performed across 6 transport modes: automobile, bicycle,  
36 bus, ferry, rail and walking. These encompassed approximately 3000 individual trips where  
37 UFP concentrations were measured. After weighting mean UFP concentrations by the  
38 number of trips in which they were collected, we found overall mean UFP concentrations of  
39  $3.4, 4.2, 4.5, 4.7, 4.9$  and  $5.7 \times 10^4$  particles  $\text{cm}^{-3}$  for the bicycle, bus, automobile, rail,  
40 walking and ferry modes, respectively. The mean concentration inside automobiles travelling  
41 through tunnels was  $3.0 \times 10^5$  particles  $\text{cm}^{-3}$ .

42

43 While the mean concentrations were indicative of general trends, we found that the  
44 determinants of exposure (meteorology, traffic parameters, route, fuel type, exhaust treatment  
45 technologies, cabin ventilation, filtration, deposition, UFP penetration) exhibited marked  
46 variability and mode-dependence, such that it is not necessarily appropriate to rank modes in  
47 order of exposure without detailed consideration of these factors. Ten in-transit health effects  
48 studies have been conducted and their results indicate that UFP exposure during commuting  
49 can elicit acute effects in both healthy and health-compromised individuals. We suggest that  
50 future work should focus on further defining the contribution of in-transit UFP exposure to

51 total UFP exposure, exploring its specific health effects and investigating exposures in the  
52 developing world.

53

54 **Keywords:** air pollution; transport modes; acute health effects; travel; public transport

55

## 56 **1. INTRODUCTION**

57 The study of commuter exposure to traffic-related air pollutants is not a particularly new field  
58 of research. Among the first researchers to recognise its significance was Professor Arie  
59 Haagen-Smit, who is best-known for his pioneering and enduring work related to  
60 characterising photochemical smog and ozone. In 1966, he performed a series of carbon  
61 monoxide measurements on heavily trafficked Los Angeles roads (Flachsbart, 2007; Haagen-  
62 Smit, 1966). Given population growth and increased motor vehicle use since that time,  
63 coupled with the high degree of proximity to vehicle emissions when commuting, the issue of  
64 ‘in-transit’ exposure to air pollutants is of equal if not greater relevance 45 years later.

65

66 Previous reviews of in-transit pollutant exposure, of which there are few, have focussed on  
67 CO inside vehicles (El-Fadel and Abi-Esber, 2009), particle mass concentrations and  
68 composition in metro (subway) systems (Nieuwenhuijsen et al., 2007) and various pollutants  
69 in multiple transport modes (Weisel, 2001). Only the work of Kaur et al. (2007) included a  
70 review of ultrafine (<100 nm) particle (UFP) concentrations in several transport modes.

71

72 At present, although gaseous pollutants are still the focus of numerous in-transit exposure  
73 studies, UFPs are beginning to attract significant attention. They are produced in large  
74 quantities by fuel combustion, and have been identified as a causal component of various  
75 negative health effects in humans (Knol et al., 2009; Hoek et al., 2010). UFPs typically

76 constitute ~90% or more of particle number count (PNC) in areas influenced by vehicle  
77 emissions (Morawska et al., 2008), and we use UFP to describe PNC throughout this article.

78

79 The primary aim of this review is to distil the results of work performed to-date in order to  
80 improve understanding of the measurement, characteristics and determinants of in-transit  
81 exposure to UFPs, prior to a discussion of gaps in knowledge and suggestions for future  
82 research. Here, we extend the work of Kaur et al. (2007) by confining our focus to UFPs and  
83 incorporating the substantial body of relevant work that has appeared in the 4 years since its  
84 publication, which now constitutes the large majority of available literature. Like Kaur et al.  
85 (2007), we restrict our investigation to UFP exposure concentrations, rather than average or  
86 integrated exposure for a given time period. We note that dose assessment, which is a  
87 complementary yet distinct concept to that of exposure (Ott, 1985), is not the main focus of  
88 this review.

89

90 This review begins with an overview of the nature of commuter travel prior to a description  
91 of the general characteristics of UFPs. This is followed by a detailed analysis of in-transit  
92 UFP exposure studies, a discussion of determinant factors, health effects, and suggestions for  
93 future research.

94

### 95 **1.1 Commuting in modern society**

96 The nature of modern society in many countries both affords and expects a high degree of  
97 personal mobility. Time-activity patterns define how people apportion their time across a  
98 range of environments, and are a keystone of effective exposure analyses. Time-activity  
99 pattern studies of varying magnitude performed in different regions have reported that time  
100 spent in-transit typically constitutes between about 5 and 10% of the day (Klepeis et al.,

101 2001), depending on location. The transport microenvironment(s) within which this time is  
102 spent varies more substantially between regions than the occupancy time, and has a greater  
103 dependence on local factors, such as the availability and desirability of various transport  
104 options.

105

106 In general, there are scant 24 hour time-activity pattern data for developing countries.

107 Saksena et al. (2007) reported that time spent travelling among 4311 Delhi residents ranged  
108 from 0.8 to 10% of the day, and varied markedly depending on age, sex and occupation, as  
109 did the mode of transport used. It is likely that the time-activity patterns of people in rural  
110 areas differ significantly from those of their urban counterparts.

111

## 112 **1.2 Children's and adult's travel choices**

113 Children are particularly susceptible to negative health effects caused by exposure to air  
114 pollutants (Gauderman et al., 2004; Brugge et al., 2007; Ashmore and Dimitroulopoulou,  
115 2009), and many millions are required to commute between home and school each weekday.  
116 The choice of which transport mode school children utilise is normally at the discretion of  
117 others. Whilst children and young people have been reported to possess informed and  
118 responsible opinions regarding transport choices and a clear ideal towards cycling and  
119 walking, their parent's choices are guided primarily by safety concerns, and place substantial  
120 reliance on private automobiles (Lorenc et al., 2008).

121

122 Unlike children, adults generally make their own travel choices. A recent survey of 745  
123 employed adults in Queensland, Australia, found that while about half of respondents felt that  
124 exposure to air pollutants in-transit negatively affected their overall health and increased their  
125 risk of cardiovascular disease, only 13% indicated that exposure to pollutants was a barrier to

126 their adoption of walking or cycling to work, and that other factors were more responsible for  
127 their high level (82%) of dependence on private transport (Badland and Duncan, 2009).  
128 Furthermore, Badland and Duncan (2009) found that adults who were better educated and  
129 lived in urban areas were most cognisant of the negative health effects of air pollutant  
130 exposure during transit. Marshall et al. (2009) reported that the optimum balance between  
131 high walkability and low pollution was identified sporadically and typically in higher income  
132 neighbourhoods in urban Vancouver (Marshall et al., 2009). Evidently, there may be a  
133 significant socio-economic component involved in air pollution exposure during transit,  
134 particularly for active transport modes, and this may reflect wider socio-economic and  
135 environmental inequalities reported for several traffic pollutants (Marshall, 2008; Tonne et  
136 al., 2008; Su et al., 2009). It should be noted that both children and adults in developing  
137 countries are unlikely to be afforded the luxury of a travel choice, *per se*, and a relatively  
138 high degree of dependency on walking and public transport may result from this (Saksena et  
139 al., 2007).

140

## 141 **2. CHARACTERISTICS OF ULTRAFINE PARTICLES**

### 142 **2.1 General**

143 UFP concentrations reflect the contribution of anthropogenic processes to a pre-existing  
144 background concentration (Morawska et al., 2008). Background concentrations are ascribed  
145 to natural processes, such that in most environments free from the immediate influence of  
146 anthropogenic activities, UFPs are present and their concentrations readily measured.  
147 Despite the numerous natural sources of UFPs, vehicular fossil fuel combustion has  
148 repeatedly been shown to be their dominant source in urban areas, with heavy-duty diesel  
149 powered vehicles making a disproportionately large contribution to UFP concentrations  
150 (Morawska et al., 2008).

151

152 An important distinction is between primary and secondary UFPs. The primary variety are  
153 emitted from their source as particles, whilst secondary particles are formed following  
154 homogenous nucleation of gases (Koutrakis and Sioutas, 1996; Jacobson, 2002). This occurs  
155 when a gas, or gases, nucleate in the absence of a pre-existing surface (Jacobson, 2002).

156

157 UFPs from vehicles can be emitted as primary particles or generated as a secondary aerosol,  
158 often following homogenous nucleation of  $\text{SO}_2$ ,  $\text{NH}_3$  and  $\text{NO}_x$  into  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  
159  $\text{NO}_3^-$  (Koutrakis and Sioutas, 1996; Morawska et al., 2008). The ratio of primary to  
160 secondary particles varies substantially according to fuel type and operating and  
161 environmental conditions, but nucleation mode particles can comprise approximately 90% or  
162 more of UFPs in diesel exhaust (Kittelson, 1998). However, more recent research indicates  
163 that the number of nucleation mode particles in diesel exhaust can be reduced to 40-50%  
164 when ultra-low sulphur diesel fuel is used (Ristovski et al., 2006), which is more  
165 representative of modern vehicle fleets in many countries.

166

## 167 **2.2 Typical concentrations**

168 Morawska et al. (2008) performed a meta-analysis of 71 UFP studies performed across a  
169 diverse range of environments. They found mean concentrations of 2.6, 4.8, 7.3, 10.8, 42.1,  
170 48.2, 71.5 and  $167.7 \times 10^3$  particles  $\text{cm}^{-3}$  for clean background, rural, urban background,  
171 urban, street canyon, roadside, on-road and tunnel environments, respectively. This indicates  
172 that greater proximity to vehicles is associated with increased UFP concentrations, and  
173 underscores their importance as a UFP source.

174



175 **2.3 Health significance**

176 Once inhaled, UFPs can reach with the alveolar region of the human lung with greater  
177 efficiency than larger particles due to their smaller size, and can deposit in alveoli with  
178 greater efficiency as a consequence of their rapid diffusion (Daigle et al., 2003; Phalen et al.,  
179 2006; Frampton, 2007). Due to their content of reactive oxygen species (ROS) and large  
180 combined surface area, UFPs from vehicle emissions have the potential to damage pulmonary  
181 cells (Delfino et al., 2005). Transition metal components in UFPs are believed to play a role  
182 in producing ROS, along with pro-oxidative organic hydrocarbons (Li et al., 2003).  
183 Additionally, target cells, such as airway epithelial cells and macrophages, produce ROS  
184 during biologically catalysed redox reactions occurring in the mitochondria in response to  
185 UFP uptake (Li et al., 2003; Nel, 2005). UFPs can evade alveolar macrophage clearance from  
186 the lung and enter lung cells, the interstitium and possibly the vascular bed (Geiser et al.,  
187 2005; Frampton, 2007), and can travel from the lung via blood and lymphatic circulation to  
188 other organs (Elder et al., 2006; Samet et al, 2009). UFPs are more proatherogenic than larger  
189 particles due to their greater bioavailability of reactive compounds, content of redox-active  
190 compounds, high number concentration and increased lung retention (Aruajo et al., 2009).  
191  
192 Epidemiologic investigations of UFPs have been constrained by the scarcity of UFP  
193 monitoring sites and the substantial spatial heterogeneity of concentrations (Brook et al.,  
194 2010). Studies performed to-date in Erfurt, Germany, have indicated that UFP effects on  
195 daily mortality may be of comparable magnitude to, yet independent of, those of fine  
196 particles (i.e. PM<sub>2.5</sub>), albeit with greater time lag between UFP concentrations and their  
197 effects (Wichmann and Peters, 2000). More recent results from the same long-term study  
198 have shown statistically significant associations between UFP concentrations and both total  
199 and cardio-respiratory daily mortality with a four day lag period (Stölzel et al., 2007).

200 Interestingly, this study found no association between PM<sub>2.5</sub> mass concentration and  
201 mortality. Mortality from stroke amongst aged residents of Helsinki during summer was  
202 associated with both PM<sub>2.5</sub> and UFP concentrations on the previous day, and effects were  
203 mostly independent (Kettunen et al., 2007).

204

205 The effects of UFP concentration on mortality and morbidity due to various causes are less  
206 well understood than those of larger particles. A recent elicitation of European experts found  
207 that short-term UFP exposure was rated to variously possess a medium to very high  
208 likelihood of causality for all-cause mortality, and a low to high likelihood for cardiovascular  
209 and respiratory hospital admissions (Knol et al., 2009). Long-term UFP exposure was  
210 generally rated to possess a slightly lower likelihood of causality for all-cause mortality,  
211 owing mainly to the lack of long-term studies (Knol et al., 2009). The same group of experts  
212 estimated that a permanent decrease in annual average UFP concentration of 1000 particles  
213 cm<sup>-3</sup> across Europe would lead to median decreases of 0.3%, 0.2% and 0.16% in all-cause  
214 mortality, and cardiovascular and respiratory hospital admissions, respectively (Hoek et al.,  
215 2010). The relatively small number of epidemiological studies (14) and absence of long-term  
216 studies, however, resulted in most experts indicating a substantial degree of uncertainty in  
217 their estimates (Hoek et al., 2010).

218

### 219 **3. STUDIES OF UFP CONCENTRATION IN TRANSPORT MODES**

#### 220 **3.1 Methods**

221 We searched combinations of the terms “ultrafine particle”, “transport mode”, “commuter”,  
222 “exposure” “public transport”, “microenvironment”, “vehicle”, “car”, “automobile”, “bus”,  
223 “cyclist”, “bicycle”, “train”, “metro”, “subway” on PubMed, ISI Web of Knowledge and  
224 Google Scholar until October, 2010. The reference lists of studies identified by this method

225 were reviewed for links to additional literature. Furthermore, the authors' own literature  
226 collections were utilized.

227

228 We restricted our investigation to studies that presented numeric values of UFP  
229 concentrations, and identified 47 that fulfilled this requirement. Tables S1-S7 in the  
230 Supplementary Information file contain detailed information on the various studies. These  
231 spanned 6 distinct transport modes: automobile, bus, cycling, ferry, rail and walking. Some  
232 studies dealt with multiple transport modes, whilst others focussed on a single mode. Of the  
233 studies we identified, only 7/47 (15%) had previously been reviewed by Kaur et al. (2007),  
234 which highlights the rapid progression of research related to in-transit UFP exposure since  
235 publication of their work.

236

237 The mean concentrations extracted from the studies identified were weighted by the  
238 corresponding number of trips taken, and overall trip-weighted mean UFP exposure  
239 concentrations were calculated for each transport mode (see tables S1-S7 in the  
240 Supplementary Information file). The overwhelming majority of studies (93%) reported the  
241 number of trips associated with a given mean; the means reported by those that did not report  
242 trip number were weighted by a conservative factor of 1 trip. Most studies reported  
243 arithmetic mean UFP concentration, while several reported geometric mean and one gave  
244 median values. Where possible, data were disaggregated to permit analyses of the effect of  
245 variables such as fuel type, presence of exhaust-treatment devices and route.

246

247 Given the range of conditions under which they were collected, we did not assess the  
248 statistical significance of differences in measured mean UFP concentrations between modes,

249 and instead sought to identify general trends in the data. This is discussed further in sections  
250 4 and 5.

251

### 252 **3.2 Results**

253 Across all modes, we identified approximately 3000 individual trips where UFP  
254 measurements were performed. There was an uneven distribution of measurement trips; very  
255 few have been performed in ferry (13) and rail (49) modes, while a substantial number have  
256 been undertaken in bus (505), walking (524), cycling (599) and automobile (1310) modes.  
257 The automobile mode was split into non-tunnel (977) and tunnel (333) trips, as previous  
258 results indicate that tunnels are a discrete UFP exposure microenvironment distinct from open  
259 air roadways (Kaminsky et al., 2009; Knibbs et al., 2010).

260

261 Figure 1 shows the trip-weighted mean UFP concentrations for each mode, and the number of  
262 trips on which they were based. Error bars indicate the trip-weighted standard deviation  
263 (Bland and Kerry, 1998). The range of mean UFP concentrations spanned one order of  
264 magnitude, with the lowest measured whilst cycling and the highest in automobiles during  
265 tunnel travel;  $3.4 \times 10^4$  (s.d. =  $1.8 \times 10^4$ ) and  $3.0 \times 10^5$  (s.d. =  $2.6 \times 10^5$ ) particles  $\text{cm}^{-3}$ ,  
266 respectively. Means and standard deviations calculated for the automobile (non-tunnel), bus,  
267 ferry, rail and walk modes were 4.5 (3.3), 4.2 (3.1), 5.7 (0.5), 4.7 (4.1) and  $4.9 (3.2) \times 10^4$   
268 particles  $\text{cm}^{-3}$ , respectively.

269

270 FIGURE 1 TO BE INSERTED HERE

271

### 272 **4. COMPARISON BETWEEN MODES**

273 Considering the diversity of studies from which they were drawn, the trip-weighted  
274 concentrations measured in automobile (non-tunnel), cycle, bus, rail and walking modes  
275 exhibited notable coherence, with a maximum to minimum ratio (walk:cycle) of 1.5. A  
276 limited number of studies that measured concentrations in different modes simultaneously or  
277 near-simultaneously have been reported, and Briggs et al. (2008) found a walk:automobile  
278 ratio of 1.4, which is higher than the value of 1.1 presented here. Boogaard et al. (2009)  
279 found an automobile:cycle ratio of 1.05, whilst we found a value of 1.3, which was also  
280 higher than the value of approximately 1.0 reported by Int-Panis et al. (2010).

281

282 While the above studies highlighted the relative concentrations encountered in each mode in  
283 the absence of bias due to fluctuating UFP concentrations, observed inter-mode contrasts  
284 were specific to the conditions of the study (e.g. the ventilation settings in an automobile, or  
285 the proximity to traffic on a bike route) and are should therefore not be extrapolated beyond  
286 the conditions under which they were collected without appropriate caution.

287

288 In studies that measured UFP concentrations in multiple modes non-simultaneously, the  
289 mode in which highest concentrations were recorded vacillated between automobiles and  
290 buses, whilst those in other modes were markedly lower (Levy et al., 2002; Kaur et al.,  
291 2005b; Weichenthal et al., 2008; Cattaneo et al., 2009; Kaur and Nieuwenhuijsen, 2009;  
292 Pattinson, 2009; Shrestha, 2009; Knibbs and de Dear, 2010). It is therefore noteworthy that  
293 our analysis found that UFP concentrations in buses and automobiles (non-tunnel) were  
294 relatively low. We pooled a large number of reported UFP measurements performed under a  
295 wide range of conditions, and while the ability to differentiate the observed differences is  
296 limited by the level of detail given by the various studies, such an approach is indicative of  
297 mean values and general trends. However, the mode in which highest exposures are

298 experienced depends strongly on the determinant factors discussed in the following two  
299 sections, and generalisation of results may be of limited value (Int-Panis et al., 2010); that is,  
300 within mode variability is likely to be substantial.

301

## 302 **5. DETERMINANTS OF UFP CONCENTRATION IN-TRANSIT**

303 Despite the convenience it may provide, it is not necessarily appropriate to rank transport  
304 modes in order of UFP exposure without certain caveats. For example, figure 1 shows the  
305 trip-weighted mean UFP concentration in an automobile is higher than the equivalent for  
306 cycling. However, an occupant of a relatively air-tight automobile in which air is recirculated  
307 and filtered will likely experience markedly lower exposure concentrations than a cyclist on a  
308 high traffic route. Disentangling the relative roles of determinant factors, their interactions  
309 and variability in each mode is a key element required to advance understanding of in-transit  
310 UFP exposure. The data reviewed here suggest that while the relationship between UFP  
311 concentration and its determinants is often mode-dependent, exposure in all mode types is the  
312 result of interplay between multiple factors. These can be viewed as comprising two stages:  
313 the first determines the outdoor or on-road UFP concentration, and the second determines  
314 what proportion of this is able to come into contact with a commuter. These factors are  
315 addressed, in turn, in the following sections.

316

### 317 **5.1 Meteorological variables**

318 Temperature has been variously reported to be positively and negatively correlated with UFP  
319 concentrations, although in vehicle-dominated areas the correlation is more likely to be  
320 negative due to condensation of volatile compounds in emissions (Morawska et al., 2008).  
321 In-transit studies that assessed this relationship uniformly found a negative correlation  
322 between temperature and UFP concentration (Krausse and Mardaljevic, 2005; Vinzents et al.,

323 2005; Thai et al., 2008; Weichenthal et al., 2008; Kaur and Nieuwenhuijsen, 2009; Pattinson,  
324 2009; Laumbach et al., 2010). Among studies that reported correlation coefficients, those  
325 measured for cycling studies (Vinzents et al., 2005; Thai, 2008) were quite high (-0.62 and -  
326 0.76, respectively). Multi-mode and automobile studies reported correlations of -0.77 and -  
327 0.37, respectively (Kaur and Nieuwenhuijsen, 2009; Laumbach et al., 2010).

328

329 Wind speed, which affects dilution and transport of vehicle emissions, was also found to be  
330 negatively correlated with UFP concentration in-transit (Krausse and Mardaljevic, 2005;  
331 Vinzents et al., 2005; Briggs et al., 2008; Thai et al., 2008; Weichenthal et al., 2008; Kaur  
332 and Nieuwenhuijsen, 2009; Pattinson, 2009; Srestha, 2009; Knibbs and de Dear, 2010),  
333 which is in agreement with results reported for various outdoor locations (Morawska et al.,  
334 2008). However, the results were not always statistically significant, indicating that  
335 temperature may be more consistently and strongly correlated with UFP concentrations.  
336 Correlations observed for active transport modes were -0.20 (walk), -0.52 (cycle) and -0.81  
337 (cycle) (Briggs et al., 2008; Vinzents et al., 2005; Thai et al., 2008). Multi-mode and  
338 automobile studies reported correlations of -0.14 to -0.49 (Briggs et al., 2008; Kaur and  
339 Nieuwenhuijsen, 2009; Knibbs and de Dear, 2010).

340

341 For both temperature and wind speed, stronger correlations were generally observed for  
342 cycling compared to non-active modes, perhaps reflecting reduced influence of other factors  
343 on exposure concentrations encountered when cycling (and walking). The strength of the  
344 association between UFP concentration and both temperature and wind speed appears to be  
345 mode and location-dependent, and its variability is not well characterised.

346

347 While temperature and wind speed are the most frequently reported, other meteorological  
348 parameters may affect UFP concentration. The depth of the mixed layer within the  
349 atmosphere was found to be negatively correlated with in-transit UFP concentration  
350 (Weichenthal et al., 2008), which reflects the tendency of a shallow mixed layer to  
351 concentrate pollutants.

352

## 353 **5.2 Traffic volume and composition**

354 Very few studies have reported the relationship between traffic volume and in-transit UFP  
355 concentrations. Fewer still have examined the effect of traffic composition (i.e. gasoline  
356 vehicles, diesel vehicles). Briggs et al. (2008) observed statistically significant correlations  
357 between car and truck density and UFP concentrations encountered while walking ( $r = 0.41$   
358 to  $0.48$ ) or in an automobile ( $r = 0.43 - 0.47$ ) in London. In their London-based study, Kaur  
359 and Nieuwenhuijsen (2009) similarly found a significant correlation ( $\rho = 0.27$ ) between total  
360 traffic count and UFP concentrations in automobile, bus, cycle, taxi and walking modes.  
361 Krausse and Mardaljevic (2005) reported road link end description (e.g. signal, left turn, right  
362 turn etc.) was a significant determinant of total UFP exposure of car occupants. On-road  
363 studies have shown strong associations ( $R^2 \sim 0.85$ ) between heavy diesel traffic volume and  
364 UFP concentrations (Fruin et al., 2008; Knibbs et al., 2009b). Other studies have reported  
365 more qualitative assessments of traffic effects; for example, that mean in-transit UFP  
366 concentrations increased on high traffic routes and vice-versa (Zhu et al., 2007; Thai et al.,  
367 2008; Strak et al., 2010; Zuurbier et al., 2010).

368

369 Vehicle emissions are the dominant source of UFPs in urban areas, and heavy diesel vehicles  
370 make a contribution that is disproportionate to their volume (Morawska et al., 2008).

371 Coupled with the limited but consistent findings of in-transit studies, this suggests that traffic



372 parameters (volume, density) and composition (gasoline vehicles, heavy diesel vehicles) are  
373 an important determinant of in-transit UFP exposure. It should be considered, however, that  
374 effects are likely to depend on mode, and that short-term traffic patterns not represented in  
375 hourly or daily average data, such as the impact of passing traffic, may be important (Fruin et  
376 al., 2008; Boogaard et al., 2009).

377

### 378 **5.3 Route choice: active transport modes**

379 There are generally fewer mode-specific variables that may affect pedestrians and cyclists  
380 compared to other transport modes; that is, traffic and meteorological conditions may be of  
381 greater importance as determinants. Most cycling studies were performed on or proximate to  
382 major urban roads, however, some studies compared measurements on high and low traffic  
383 routes, with the latter typically comprised of a dedicated cycle path. Separating the data into  
384 these two categories revealed that 18% of trips were undertaken on low traffic routes, and  
385 mean UFP concentrations were  $2.6 \times 10^4$  particles  $\text{cm}^{-3}$ . The mean for high traffic routes was  
386  $3.5 \times 10^4$  particles  $\text{cm}^{-3}$ , suggesting that route selection, within the context of the few studies  
387 to address it, can affect cyclist UFP exposure (Pattinson, 2009; Strak et al., 2010; Zuurbier et  
388 al., 2010).

389

390 Route choice, as a proxy for traffic volume, is likely to be an important determinant of  
391 exposure (McCreanor et al., 2007; Hertel et al., 2008), and personal factors (e.g. walking or  
392 cycling patterns) may also exert an effect (Kaur et al., 2007). Microscale variations in UFP  
393 concentration proximate to roadways may result in higher exposures on the road side of the  
394 sidewalk/footpath compared to the building side (Kaur et al., 2005a). Also, the effect of  
395 roadway factors on pollutant dispersion (i.e. whether open to the environment or prone to trap  
396 pollutants due to geometry of urban canyons) has been shown to be a statistically significant

397 determinant of UFP exposure concentrations encountered when walking (Briggs et al., 2008).  
398 Further work focussed on evaluating the effects of these local and microscale route  
399 phenomena on UFP exposure is required.

400

#### 401 **5.4 Cabin ventilation**

402 Ventilation rates, whether driven by fans, natural leakage or open windows (Ott et al., 2008;  
403 Knibbs et al., 2009a), describe how rapidly outdoor air is capable of entering passenger  
404 cabins. Evidence suggests that ventilation is a key determinant of in-cabin UFP  
405 concentrations in automobiles, buses (Hammond et al., 2007; Rim et al., 2008; Knibbs and de  
406 Dear, 2010; Zhang and Zhu, 2010), ferries (Hill et al., 2007; Knibbs and de Dear, 2010) and  
407 rail modes (Hill et al., 2007; Cheng et al., 2009; Knibbs and de Dear, 2010). Quantitative  
408 studies support these observations, but are scarce and limited to automobiles (Xu and Zhu,  
409 2009; Knibbs et al., 2010).

410

411 Knibbs et al. (2009a) found that air exchange increased linearly with vehicle speed in a group  
412 of six test automobiles operating under four distinct ventilation settings, which was in  
413 agreement with results obtained by Ott et al. (2008) based on tests performed in four vehicles.  
414 Knibbs et al. (2009b; 2010) found that the primary determinant of on-road UFP concentration  
415 in a tunnel bore was hourly heavy diesel vehicle volume ( $R^2 = 0.87$ ), and that cabin  
416 ventilation rates explained 81% of the variation in the proportion of on-road UFPs reaching  
417 the occupants of 5 automobiles. The proportion reaching the cabin varied from 0.08  
418 (recirculation) to ~1.0 (non-recirculation) depending on vehicle and ventilation setting. Thus,  
419 ventilation rates controlled the extent to which in-cabin exposure concentrations reflected on-  
420 road levels in the tunnel bore, which were largely determined by heavy diesel vehicle  
421 volume. Xu and Zhu (2009) reported that cabin ventilation and leakage were predominant

422 factors in their model-based analyses of variables affecting in-cabin/on-road (I/O) ratios, and  
423 explained up to ~60% of on-road UFP ingress. I/O ratios when windows are open can reach  
424 1 due to higher air exchange, and such conditions may also occur when windows are closed  
425 but ventilation fan settings are high (Ott et al., 2008; Knibbs et al., 2009a).

426

427 Some investigators have successfully performed in-cabin UFP size distribution measurements  
428 during transit in automobiles (Zhu et al., 2007) and buses (Zhang and Zhu, 2010). These  
429 studies have shown that while in-cabin particle size distributions follow the general shape of  
430 those on-roads, the ability of on-road particles to reach the cabin is dependent on particle size  
431 and ventilation settings (Zhu et al., 2007). Particle penetration is discussed in section 5.6.

432

### 433 **5.5 Filtration**

434 Where a vehicle is fitted with a cabin air filter, its efficiency is a key determinant of what  
435 proportion of on-road UFPs reach the cabin, and efficiency varies substantially amongst the  
436 filters available. Standard automobile cabin filters afford single-pass UFP reductions of  
437 between approximately 30 and 60% (Pui et al., 2008; Qi et al., 2008), while this can  
438 increased by employing more advanced filtration technologies (Burtscher et al., 2008). It  
439 should be noted that filtration efficiency is affected by the ventilation rate; as filter face  
440 velocity increases with mechanical or natural ventilation rates, filtration efficiency decreases  
441 due to the reduced time available for particle diffusion inside the filter (Pui et al., 2008; Qi et  
442 al., 2008). When air is recirculated in an automobile, Qi et al. (2008) found that UFP  
443 concentrations decayed most rapidly in a vehicle capable of filtering recirculated air (single  
444 pass efficiency = 46%) than in a vehicle lacking this feature, where UFP removal efficiency  
445 without a filter was 27% per recirculation of cabin air. In the former and latter cases, on-road  
446 UFP concentrations were reduced to those typical of an office building (4000 particles cm<sup>-3</sup>)

447 in 3 minutes and 9-10 minutes, respectively, indicating the value of recirculation as a simple  
448 exposure minimisation mechanism. However, some older, less-airtight vehicles are  
449 characterised by outdoor air exchange rates up to  $47 \text{ hr}^{-1}$  when air is recirculated (Knibbs et  
450 al., 2009a), and the benefit of recirculation in such cases can be substantially diminished  
451 (Knibbs et al., 2010).

452

### 453 **5.6 UFP penetration and deposition**

454 The penetration of UFPs through automobile envelopes is dependent on their size, the  
455 number and geometry of cracks, and the pressure difference across these and other ingress  
456 pathways (Xu et al., 2010). A recent study reported that penetration efficiency close to 100%  
457 was observed for diesel exhaust particles between 100 and 287 nm, and declined to ~70% for  
458 10 nm particles due to diffusion; although penetration of 10 nm particles increased to ~90%  
459 when pressure differentials reached 200 Pa (Xu et al., 2010). No difference was observed in  
460 penetration efficiency amongst different materials.

461

462 Given the high surface to volume ratios of many automobile cabins, deposition can be an  
463 important UFP removal mechanism, especially under low ventilation conditions (Gong et al.,  
464 2009). Gong et al. (2009) found in-cabin deposition rates in automobiles exceed those of  
465 indoor environments by a factor of 3 to 20.

466

467 Studies describing UFP filtration, penetration and deposition in bus and rail modes are scarce  
468 and the limited data to-date is strongly skewed towards automobiles. Future studies  
469 addressing this knowledge gap will be of considerable value.

470

### 471 **5.7 Fuel type and presence of an emission control device**

472 *Automobile*: The effect of fuel type on UFP concentration in automobiles was assessed by  
473 Zuurbier et al. (2010), who found no significant difference in mean levels in diesel and  
474 gasoline-powered vehicles (diesel:gasoline concentration ratio = 0.96) based on 14  
475 simultaneous trips under a standard ventilation setting. Their study focussed only on newer  
476 vehicles (< 6 months) and its relevance to the wider passenger vehicle fleet is unknown.  
477 Additionally, it is difficult to separate the effects of fuel type from those due to differences in  
478 ventilation under a standard setting between vehicles of different manufacturer (e.g. Knibbs  
479 et al., 2009a). Further studies involving test vehicle groups more representative of the  
480 heterogeneity present in wider vehicle fleets are required.

481

482 *Bus*: Due to their frequency of door opening and the ‘stop-start’ nature in which they travel,  
483 buses have a tendency to self-pollute (Behrentz et al., 2004; Hill et al., 2005; Rim et al.,  
484 2008; Liu et al., 2010; Zhang and Zhu, 2010; Zuurbier et al., 2010). Accordingly, the  
485 variables most frequently reported by UFP exposure studies were fuel type and the presence  
486 of an exhaust or crankcase emission control device. We therefore disaggregated bus trips  
487 into 8 categories: diesel, biodiesel, compressed natural gas (CNG), electric, diesel with  
488 oxidation catalyst (DOC), diesel with diesel particulate filter (DPF), diesel with crankcase  
489 filtration system (CFS), and diesel with combined control (i.e. any combination of two or  
490 more of DOC, DPF, CFS and ultra low sulphur diesel). About 70% of trips were performed  
491 in diesel buses, with the remainder approximately evenly distributed across the other  
492 categories. Five percent of bus trips (26/505) were excluded due to lack of detailed data on  
493 fuel type or control device.

494

495 Figure 2 shows the trip-weighted mean UFP concentrations for each category. The lowest  
496 mean ( $1.7 \times 10^4$  particles  $\text{cm}^{-3}$ ;  $\text{SD} = 0.8 \times 10^4$ ) was recorded in CNG-powered buses, and the

497 highest ( $4.9 \times 10^4$  particles  $\text{cm}^{-3}$ ;  $\text{SD} = 2.6 \times 10^4$ ) was measured in diesel buses fitted with a  
498 CFS, although the latter result was based on a very limited number of trips (13). A similar  
499 mean was recorded in diesel buses with no control device ( $4.8 \times 10^4$  particles  $\text{cm}^{-3}$ ;  $\text{SD} = 3.2$   
500  $\times 10^4$ ). Means and standard deviations calculated for the biodiesel, combined control, DPF,  
501 DOC and electric categories were 1.7 (-), 2.0 (1.8), 2.4 (0.9), 2.8 (2.0) and 2.9 (0.8)  $\times 10^4$   
502 particles  $\text{cm}^{-3}$ , respectively. With the exception of the electric bus category, lowest  
503 concentrations were measured in buses powered by alternative fuels. Concentrations inside  
504 diesel-powered buses were generally lower when fitted with an emission control device.

505

506 FIGURE 2 TO BE INSERTED HERE

507

508 Differentiating the effects of self-pollution from those of other factors on in-bus UFP  
509 concentrations is challenging. Previous work has shown that self-pollution can be the  
510 dominant source of vehicle emissions in the cabin when windows are closed, and constituted  
511 0.01 to 0.3% of air in the cabins of 1975 through 2002 model school buses (Behrentz et al.,  
512 2004). Liu et al. (2010) found that self-pollution contributed an overall average of  $1.8 \times 10^4$   
513 particles  $\text{cm}^{-3}$  in two school buses (2000 and 2003 model); the average contribution when  
514 windows were closed ( $1.0 \times 10^4$  particles  $\text{cm}^{-3}$ ) was less than that when they were open ( $2.6 \times$   
515  $10^4$  particles  $\text{cm}^{-3}$ ). However, this trend was not in keeping with their results for other  
516 measured pollutants, and was attributed to UFP fluctuations due to unidentified non-vehicle  
517 sources on the low-traffic routes they studied. Generally, if on-road concentrations are low  
518 relative to those in-cabin, open windows will dilute self-pollution (Liu et al., 2010). The  
519 reverse can exacerbate its effects.

520

521 The relatively small number of trips taken in most categories we analysed and the lack of  
522 specific information regarding other possible determinants limits the conclusions that can be  
523 drawn, and precluded detailed statistical analyses. However, the results generally suggest  
524 that UFP concentrations are greatest in diesel-powered buses, and that reductions may be  
525 possible through use of alternative fuels or emission control devices, with best results  
526 achieved for diesel buses when two or more of the latter are combined.

527

528 *Rail:* In most rail studies we identified, trips were undertaken in vehicles driven by electricity.  
529 About 29% of trips were taken in diesel-powered trains, and the weighted mean UFP  
530 concentration during these was  $9.0 \times 10^4$  particles  $\text{cm}^{-3}$ . The mean during travel in electric-  
531 powered vehicles was  $3.0 \times 10^4$  particles  $\text{cm}^{-3}$ . Based on the limited data available, the power  
532 source of the rail vehicle therefore appears to affect UFP exposure concentrations. Moreover,  
533 in diesel trains, the position of the locomotive relative to the passenger compartments can  
534 markedly affect UFP concentrations; when a locomotive was located in front of passenger  
535 cabins, its emission plume can reach the cabin ventilation system intake, and vice-versa (Hill  
536 et al., 2007).

537

538 There was insufficient data to investigate the effect of underground and above ground travel  
539 on UFP concentrations. Whilst there are numerous mechanical processes that can generate  
540 and resuspend particulate matter in electric-powered subway/metro systems, these are more  
541 likely to elevate levels of particle mass rather than UFP number count (Nieuwenhuijsen et al.,  
542 2007). The limited number of studies reporting UFP measurements on underground  
543 platforms tend to support this (Aarnio et al., 2005; Seaton et al., 2005; Raut et al., 2009;  
544 Cheng et al., 2009; Nystrom et al., 2010).

545

546 **6. CORRELATION WITH OTHER AIR POLLUTANTS**

547 Several in-transit studies measured UFPs and other pollutants simultaneously. A summary of  
548 these is provided in Table S8 in the Supplementary Information file. The correlation between  
549 UFP and PM<sub>2.5</sub> concentrations is generally reported to be positive, weak and not statistically  
550 significant, although stronger associations have been observed; correlation coefficients range  
551 from -0.07 to 0.69 (Aarnio et al., 2005; Kaur et al., 2005a,b; Seaton et al., 2005; McCreanor  
552 et al., 2007; Zhu et al., 2008; Berghmans et al., 2009; Boogaard et al., 2009; Knibbs and de  
553 Dear, 2010; Laumbach et al., 2010). Although correlation in the rail mode is moderate and  
554 relatively consistent across studies, in general there is no clear relationship between the  
555 strength of correlation and transport mode. The results are likely to be somewhat location-  
556 dependent, in keeping with those for outdoor environments, and the generally poor  
557 correlation reflects differences in the sources of particle number and mass and temporal  
558 scales involved in their dynamics (Morawska et al., 2008).

559  
560 Black carbon (BC) and elemental carbon (EC) are often well-correlated with UFP  
561 concentrations in urban air, given their shared provenance in vehicle emissions and the large  
562 extent to which BC and EC contribute to UFP chemical composition (Morawska et al., 2008).  
563 On-road and subway platform studies have shown very good correlation between UFPs and  
564 BC; 0.88 and 0.84, respectively (Aarnio et al., 2005; Westerdahl et al., 2005). Correlations  
565 were relatively weak in automobile and bus studies (mean = 0.1 to 0.2), although in-bus  
566 relationships were strongly dependent on window position, and mean correlation improved  
567 (mean = 0.62) when windows were kept open, which the authors ascribed to self-pollution  
568 under the closed window setting (Zhu et al., 2008; Zhang and Zhu. 2010). Very good  
569 correlations between UFPs and EC (0.70 and 0.84) have been reported in walking studies  
570 (Kaur et al., 2005a; McCreanor et al., 2007).



571

572 The correlation between UFP concentrations and those of NO<sub>x</sub> vary extensively from -0.33 to  
573 0.90, and no clear relationship with transport mode is apparent (Westerdahl et al., 2005;  
574 McCreanor et al., 2007; Zhu et al., 2008; Laumbach et al., 2010). The relationship with CO  
575 concentrations is similarly variable; -0.16 to 0.70 (Kaur et al., 2005a,b ; Westerdahl et al.,  
576 2005; McCreanor et al., 2007; Zhu et al., 2008; Laumbach et al., 2010). The specifics of the  
577 measurement location in terms of local emission sources are likely to explain the observed  
578 variation, and it is important to consider that in-transit measurements of particle and gaseous  
579 pollutants may exhibit poor temporal correlation due to the varying emission strength of  
580 proximate vehicles (Morawska et al., 2008; Zhu et al., 2008).

581

582 In summary, the relationship between in-transit UFP concentrations and those of other  
583 pollutants is generally inconsistent. Mode, location and environmental factors may all  
584 contribute to the observed variability, and the results gathered here from the limited pool of  
585 available studies require further validation in order to develop a more complete understanding  
586 of the associations. Currently, there is no data to support prediction of UFP concentrations  
587 from those of other pollutants, and such an approach is likely to be insufficient.

588

## 589 **7. RELATIONSHIP WITH FIXED SITE MONITORS**

590 Since the 1970s (Ott and Eliassen, 1973; Cortese and Spengler, 1976), numerous studies have  
591 investigated the ability of fixed site pollutant monitoring stations to estimate personal and  
592 commuter exposure. Generally, the ability of fixed site monitors to represent the substantial  
593 spatial and temporal variability of in-transit exposures has been sub-optimal, and carries with  
594 it numerous attendant limitations, the most important of which is underestimation of exposure  
595 (Kaur et al., 2007). UFPs are not a regulated pollutant, and are therefore not routinely

596 monitored outside of research studies. Some investigators have assessed the association  
597 between fixed site UFP concentrations and those measured concurrently in-transit.  
598  
599 Aarnio et al. (2005) reported good correlation ( $R^2 = 0.59$ ) between UFP concentrations in  
600 subway stations and those measured at an urban background site, while Seaton et al. (2005)  
601 found that the ratio of UFP concentrations measured on London Underground platforms to  
602 those above ground ranged from 0.38 to 0.68. These results are likely to reflect the absence  
603 of strong local UFP sources in subways (Aarnio et al., 2005). For above ground transport  
604 microenvironments, however, this is unlikely to be the case. Vinzents et al. (2005) reported a  
605 moderate correlation ( $r = 0.49$ ) between UFP measurements performed at a fixed roadside  
606 location and those measured while cycling, but found that the only significant variables in a  
607 linear mixed effects model to predict cyclist exposure were temperature and concentrations of  
608 CO ( $R^2 = 0.60$ ) and NO<sub>2</sub> ( $R^2 = 0.74$ ) measured at urban background and roadside stations,  
609 respectively. Asmi et al. (2009) found that the ratio of UFP concentration in the driver's  
610 cabin of buses to that measured at an urban background site varied from 1.2 to 6.9 and was  
611 dependent on the age of the bus, time of day and route. Zuurbier et al. (2010) systematically  
612 evaluated the relationship between bus, car and bicycle UFP exposures and urban background  
613 concentrations in Arnhem, the Netherlands. They reported median mode to background  
614 ratios of 1.6 (diesel car, electric bus) to 2.5 (diesel bus) and correlations between 0.01 (diesel  
615 bus) and 0.87 (bicycle on low-traffic route).

616  
617 The limited data available to-date indicates that fixed-site monitors may offer some ability to  
618 estimate UFP exposure of commuters in areas less affected by vehicle emissions, such as  
619 those using subways or low-traffic bike paths. However, depending on location, such persons  
620 are likely to constitute only a minor proportion of the commuting population. In the absence

621 of widespread UFP monitoring networks, the utility of routinely monitored particle and  
622 gaseous pollutants or individual UFP monitors to represent in-transit UFP exposure appears  
623 significantly constrained (Krausse and Mardaljevic, 2005; Vinzents et al., 2005).

624

## 625 **8. HEALTH EFFECTS OF IN-TRANSIT UFP EXPOSURE**

626 Studies of health effects due to commuter UFP exposure are summarised in Table S9 in the  
627 Supplementary Information file.

628

### 629 **8.1 Healthy Individuals**

630 Nystrom et al. (2010) showed that while a cellular response in the airway epithelium was not  
631 elicited, minor biological responses such as increased systemic markers of inflammation and  
632 signs of lower airway irritation were observed in 20 healthy individuals exposed to subway  
633 air (mean UFP concentration  $1.1 \times 10^4$  particles  $\text{cm}^{-3}$ ) for 2 hours while alternating between  
634 exercising on a bicycle ergometer and resting. However, road tunnel air (median UFP  
635 concentration  $1.1 \times 10^5$  particles  $\text{cm}^{-3}$ ) elicited an inflammatory response in the lower airways  
636 and elevated levels of T-lymphocytes and alveolar macrophages in brochoalveolar lavages  
637 from 16 healthy individuals who followed the same protocol (Larsson et al., 2007). The  
638 particle mass concentrations that subjects were exposed to in the two above studies were  
639 similar, while UFP and  $\text{NO}_x$  concentrations were an order of magnitude higher in the road  
640 tunnel study than in the subway study due to the presence of proximate vehicle emissions.  
641 Although it is not possible to ascribe the disparity in the results of the two studies to  
642 differences in UFP concentration alone, the results are suggestive of a causative role for UFP  
643 and  $\text{NO}_x$  in airway inflammation observed following exposure to vehicle emissions.

644

645 Thirty-eight healthy volunteers who cycled parallel to a major traffic corridor for 20 minutes  
646 (mean UFP concentration  $2.9 \times 10^4$  particles  $\text{cm}^{-3}$ ) experienced a minor increase in blood  
647 inflammatory cell distribution compared to cycling in a clean air environment, although the  
648 role of UFPs as distinct from  $\text{PM}_{2.5}$  was not clear (Jacobs et al., 2010). UFP and EC exposure  
649 in 12 healthy non-smoking individuals cycling in traffic (mean UFP concentration 2.8 to 4.1  
650  $\times 10^4$  particles  $\text{cm}^{-3}$ ) for 1 hour was weakly associated with acute effects; decreased lung  
651 function and increased exhaled NO (as a marker of airway inflammation) were observed 6  
652 hours post-exposure (Strak et al., 2010). Oxidative DNA damage observed in 15 healthy  
653 subjects was positively correlated with cumulative UFP exposure, to which 1.5 hours of  
654 cycling during rush hours (mean UFP concentration  $3.2 \times 10^4$  particles  $\text{cm}^{-3}$ ) contributed  
655 substantially and resulted in greater damage compared to indoor cycling on an ergometer  
656 (Vinzents et al., 2005). Concentrations of other pollutants ( $\text{PM}_{10}$ ,  $\text{NO}_x$ , CO) measured at  
657 fixed-sites were not associated with oxidative DNA damage.

658

659 UFP exposure resulted in modest effects among 34 healthy subjects that commuted by  
660 automobile, bus or bicycle for 2 hours (median UFP concentration 2.7 to  $4.4 \times 10^4$  particles  
661  $\text{cm}^{-3}$ ); peak expiratory flow decreased slightly and airway resistance increased immediately  
662 following exposure, and a significant increase in exhaled NO was observed 6 hours post-  
663 exposure for automobile and bus commuters, but not cyclists (Zuurbier et al., 2011).

664

665 As the respiratory minute ventilation of cyclists is 2 to 4.5 times that of automobile and bus  
666 passengers (Zuurbier et al., 2009; Int Panis et al., 2010), the potential dose of inhaled UFPs  
667 received during active transport may be significantly higher than that in non-active modes,  
668 and recent health effects studies have already begun to adopt a more dose-oriented approach  
669 to reflect this (Zuurbier et al., 2011).

670

## 671 **8.2 Health-compromised Individuals**

672 *Asthmatics:* Asthma exacerbations can be triggered due to oxidative stress and inflammation  
673 caused by UFPs in the lungs of susceptible individuals (Weichenthal et al., 2007).

674 Reductions in lung function and increased daily symptoms in asthmatics and COPD patients  
675 attributable to elevated UFP concentrations have been observed in epidemiologic studies,  
676 with more immediate effects seen first in the respiratory system, and a delayed response of  
677 cardiovascular effects (Wichmann et al., 2000; Ibalid-Mulli et al., 2002).

678

679 Consistent asymptomatic reductions in lung function (FEV<sub>1</sub>, FVC) and increases in both  
680 inflammatory biomarkers and airway acidification were observed in 60 persons with mild or  
681 moderate asthma who walked for 2 hours along a busy London street affected by diesel  
682 exhaust (median UFP concentration  $6.4 \times 10^4$  particles cm<sup>-3</sup>) (McCreanor et al., 2007). The  
683 effects were more frequently associated with UFP and EC concentrations than those of PM<sub>2.5</sub>  
684 and NO<sub>2</sub>. Significantly reduced respiratory effects were observed when subjects walked  
685 along a route less affected by traffic emissions (median UFP concentration  $1.8 \times 10^4$  particles  
686 cm<sup>-3</sup>).

687

688 Fourteen mild asthmatics exposed to road tunnel air (median UFP concentration  $2.3 \times 10^5$   
689 particles cm<sup>-3</sup>) for 2 hours while alternating between exercising on a bicycle ergometer and  
690 resting experienced no changes in bronchial responsiveness and most lung function  
691 parameters, although peak expiratory flow decreased, and minor indications of inflammation  
692 were measured in nasal lavages, but not blood samples (Larsson et al., 2010).

693

694

695 *Diabetics:* Exposure to pollutants (median UFP concentration  $4.3 \times 10^4$  particles  $\text{cm}^{-3}$ ) during  
696 1.5 to 1.8 hour automobile highway trips made by 21 type 2 diabetics was shown to elicit a  
697 decrease in high-frequency heart rate variability the day after exposure, which was more  
698 associated with the interquartile range of UFP concentration compared to those of  $\text{PM}_{2.5}$ ,  $\text{NO}_2$   
699 and CO, albeit not significantly (Laumbach et al., 2010). An increased low frequency to high  
700 frequency heart-rate variability ratio was observed immediately post-exposure that was not  
701 consistent with other observations, although confounding effects not present in the  
702 aforementioned finding may have influenced this result.

703

704 *Elderly Persons:* Nineteen elderly subjects that were exposed to unfiltered and filtered air  
705 during 2 hour automobile trips on Los Angeles freeways (mean unfiltered UFP concentration  
706  $0.78$  to  $1.1 \times 10^5$  particles  $\text{cm}^{-3}$ ) experienced a 20% decrease in the incidence of atrial ectopic  
707 heartbeats and 30% decrease in cardiopulmonary stress biomarkers under the filtered  
708 compared to the unfiltered condition (Cascio et al., 2009; Hinds et al., 2010). Other  
709 measured parameters (lung function, indicators of inflammation, blood pressure) did not vary  
710 significantly between the two conditions. The observed atrial arrhythmia was ascribed to  
711 increased intra-atrial pressure, and was associated with UFP concentrations rather than gases  
712 or particle mass (Cascio et al., 2009; Hinds et al., 2010). The significance of such events is  
713 related to their role in causing more sustained arrhythmias.

714

### 715 **8.3 Summary**

716 Commute-time exposure to traffic and attendant pollutant emissions, noise and stress has  
717 been associated with increased risk of serious adverse health effects such as myocardial  
718 infarction (Peters et al., 2004). The specific role of UFPs as a causative agent of such effects  
719 is not clear, and the findings of the limited number of health effects studies addressing

720 commuter exposure to vehicle emissions are mixed. However, some initial trends are  
721 emerging. While it is inherently difficult to separate the effects of UFPs from those other  
722 pollutants within the real-world exposure scenarios employed by the studies described above,  
723 the observed health effects were generally associated most strongly with UFP concentrations.  
724 Furthermore, the use of filtered air exposure scenarios in the Los Angeles freeway study  
725 (Cascio et al., 2009; Hinds et al., 2010) reduced particle concentration by >95% compared to  
726 the unfiltered condition but did not affect the level of gaseous pollutants, yet there was a  
727 marked difference in the cardiac effects observed between the two scenarios. The effects  
728 observed by McCreanor et al. (2007) were greater in those with moderate compared to mild  
729 asthma, and the degree to which this is true of other susceptible groups (i.e. increasing effects  
730 with increasing disease severity) is unclear. The 10 commuter health effects studies  
731 performed to-date have yielded valuable information, however, it is clear that further studies  
732 are required in order to better elucidate the role of UFPs.

733

## 734 **9. MODELLING EXPOSURE**

### 735 **9.1 Approaches employed to-date**

736 The ability to accurately model in-transit UFP exposure concentrations has numerous  
737 attractive applications in urban planning, transport and policy development. The majority of  
738 published studies that developed models employed a multivariate regression approach that  
739 incorporated meteorologic, traffic or other pollutants as independent variables (Krausse and  
740 Mardaljevic, 2005; Vinzents et al., 2005; Weichenthal et al., 2009; Boogaard et al., 2009;  
741 Kaur and Nieuwenhuijsen, 2009). Given the potential for variability in the strength of  
742 associations between the independent variables and measured UFP concentrations discussed  
743 in sections 5, the external validity of these models is unknown. However, the models were  
744 of the explanatory type, and were developed in order to assess the effect of various

745 parameters on UFP concentration measured in a specific location. Their ability to predict  
746 exposure concentrations varied from fair ( $R^2 = 0.35$ ) to very good ( $R^2 = 0.74$ ). The influence  
747 of mode-dependent parameters like ventilation were either included in a qualitative sense  
748 (e.g. ventilation setting or window position) or not included at all. This limitation was raised  
749 by both Briggs et al. (2008) and Weichenthal et al. (2008).

750

751 Several recent studies (Pui et al., 2008; Xu and Zhu, 2009; Knibbs et al., 2010) have sought  
752 to overcome the limitations described above by adopting a more mechanistic, mass-balance  
753 modelling approach for automobiles. This has been based on measurements of the effects of  
754 cabin ventilation, filtration, particle penetration or deposition on in-cabin concentrations (Qi  
755 et al., 2008; Gong et al., 2009; Knibbs et al., 2009a; Xu et al., 2010). These studies have  
756 generally shown very good results when validated with experimental data. The main  
757 limitation of such approaches is that they require the input of an initial on-road or in-cabin  
758 UFP concentration. Therefore, there is a clear need to couple models capable of predicting  
759 outdoor or on-road concentrations with those focussed on predicting what proportion of these  
760 concentrations reach occupants, and how particle dynamics will affect concentrations through  
761 time. Moreover, further refinement of models for predicting exposure in active transport  
762 modes will be of significant utility. In summary, there is both substantial need and scope for  
763 development of models capable of accurate prediction of UFP exposure concentrations in-  
764 transit. .

765

## 766 **9.2 Spatial and temporal aspects of exposure**

767 Efforts to improve understanding of the spatial and temporal nature of UFP exposures during  
768 transit have benefited greatly from the use of Global Positioning Systems (GPS) and  
769 Geographic Information Systems (GIS), usually at the measurement and analytical stages,



770 respectively. Gulliver and Briggs (2005) described the development and use of a GIS-based  
771 model for predicting exposure to PM<sub>10</sub> (particles < 10 µm) during transit, however, the  
772 application of spatial technologies to UFPs has to-date been limited to a handful of in-transit  
773 studies (Hvidberg, 2006; Thai, 2008; Berghmans et al., 2009; Boogaard et al., 2009;  
774 Pattinson, 2009; Int-Panis et al., 2010). Synchronised video recordings have been included in  
775 some studies (Kaur et al., 2006; Berghmans et al., 2009), which affords an additional  
776 perspective from which analyses can be performed.

777

778 Given the good level of spatial data quality obtainable from even the more basic mobile  
779 telephones at present, the integration of such data into exposure studies will assist data  
780 interpretation and help to form a more complete and accurate assessment of pollutant  
781 exposure and dose for large study populations (Jerrett, 2010). The appropriateness and  
782 capability of mobile telephones to record spatial data and photographs during commuting has  
783 already been established by Pooley et al. (2010), and Pattinson (2009) collected such data in  
784 addition to UFP measurements when commuting by bicycle.

785

786 Land use regression (LUR) is an application of GIS that is gaining momentum as a tool with  
787 which to predict exposure to a variety of pollutants (see Hoek et al., 2008). The utility of  
788 LUR techniques to predict UFP concentrations and spatial variability is not well-established  
789 due to absence of extensive UFP monitoring networks; other (mainly gaseous) pollutants  
790 have been the focus of most work performed to-date. However, a recent study has reported  
791 reasonable performance of LUR when applied to UFP concentrations in Amsterdam, and  
792 comparable predictive utility was observed between the LUR model for UFPs and those for  
793 other pollutants (Hoek et al., 2011). LUR is an emerging technology that will increasingly  
794 find applications in prediction of personal exposure to a range of pollutants, albeit with an

795 attendant need for validation based on measurements (Hoek et al., 2008). This highlights the  
796 need for high-quality databases of concomitant in-transit UFP and spatial measurements.

797

## 798 **10. FURTHER RESEARCH NEEDS**

### 799 **10.1 In-transit contribution to daily exposure**

800 The significance of in-transit UFP exposure is highly dependent on personal, demographic  
801 and occupational context. UFP concentrations encountered on the commute to and from  
802 work will exert much greater influence on the total daily exposure of a non-smoking office  
803 worker than a smoker or someone who experiences high occupational exposure. Likewise,  
804 the health effects of the same exposure on an adult and child are likely to vary. Without  
805 better understanding of the characteristics of 24 hour UFP exposure for numerous  
806 demographic groups, knowledge of in-transit exposure alone is of reduced utility. However,  
807 it is useful to be able to determine, for a given location, the transport mode in which highest  
808 concentrations occur and the factors that determine this. Such information has numerous  
809 valuable planning and policy applications.

810

811 A handful of studies have estimated the influence of measured in-automobile UFP  
812 concentrations on total exposure. Two were based on Los Angeles residents (Zhu et al.,  
813 2007; Fruin et al., 2008), and their estimates ranged from 10 to 50% and 33 to 45%,  
814 respectively. Wallace and Ott (2011) measured UFP concentrations in a wide range of  
815 microenvironments in two US cities and estimated the in-automobile contribution to total  
816 exposure to be 17%, which they attributed to the relatively low density of traffic and diesel  
817 trucks on the roadways they measured compared to LA. In all cases, the time spent in  
818 automobiles was assumed to be about 90 minutes per day. The applicability of the estimates  
819 reached by these studies to other regions is unknown, but they have established a range

820 within which automobile commutes in urban areas may be expected to contribute to daily  
821 UFP exposure. These estimates have flagged this topic as one requiring further investigation,  
822 preferably including several transport modes.

823

824 It is important to consider the distinction between UFP concentration and exposure (Krausse  
825 and Mardaljevic, 2005). A high concentration experienced for a brief duration can result in a  
826 lower exposure than a low concentration for a longer period. This underscores the need for  
827 both accurate time-activity pattern data across broad demographic groups and representative  
828 UFP measurements within the various microenvironments in which time is spent. Until more  
829 expansive UFP exposure studies that follow large groups of people of varying time-activity  
830 patterns are completed, the ability to discern the range of commute-time's specific  
831 contribution to total exposure is constrained.

832

### 833 **10.2 High exposure professions**

834 The magnitude of UFP exposures incurred by people whose occupation requires them to  
835 spend extended period in-transit is poorly understood. Professional drivers, bicycle couriers,  
836 police officers and other groups whose work day is constituted by long periods in transport  
837 microenvironments may all be at risk of substantially elevated exposure compared to the  
838 general population. Riediker et al. (2004) reported the negative health effects of in-vehicle  
839 PM<sub>2.5</sub> exposure on young and healthy police officers during 9 hour shifts in patrol cars.  
840 Similar studies focussed towards UFPs are required.

841

### 842 **10.3 Exposure-health effects link**

843 Various acute human health effects caused by UFP exposures have been investigated in  
844 controlled exposure studies using a range of subject groups. However, their relevance to in-

845 transit exposures is unclear. There have been precious few studies that measured the effects  
846 of in-transit exposures on health end points, and these were described in section 8. There is a  
847 significant need for further studies in this area, as they will serve to bolster the link between  
848 exposure and health effects, and this will have implications across policy, planning and  
849 public health arenas (de Nazelle and Nieuwenhuijsen, 2010). Furthermore, given the  
850 substantial variability in minute ventilation between occupants of different modes (Zuurbier  
851 et al., 2009; Int-Panis et al., 2010), the transition from an exposure to dose-oriented approach  
852 is likely to yield data of greater relevance to studies of health effects.

853

#### 854 **10.4 Data from the developing world**

855 A striking feature of the English language literature we searched is the almost complete  
856 absence of studies performed in developing regions; with the exception of only the cycling  
857 study performed in Bogota, Columbia by Fanara (2003) and cited by Kaur et al. (2007), no  
858 other studies from developing countries were identified. This shortcoming is compounded by  
859 the generally poor air quality experienced in these regions (Han and Naeher, 2006) and their  
860 large populations and urban density. The effect of this combination of factors is that very  
861 high UFP exposures are likely to occur for large numbers of people, but the magnitude of  
862 such exposures is unknown. Studies of commuter exposure to particulate mass (RSP, PM<sub>10</sub>)  
863 performed in Delhi and Hanoi have reported exceptionally high concentrations (Saksena et  
864 al., 2007; Saksena et al., 2008). Moreover, in addition to walking, the most popular modes of  
865 transport, such as bicycles, scooters, motorcycles and 3-wheelers (tuk-tuks, auto-rickshaw  
866 etc), are unlikely to afford significant protection from the emissions of proximate traffic,  
867 which can include a substantial proportion of high emitting two-stroke vehicles. There is a  
868 clear need to redress the scarcity of research in this area.

869

870 **10.5 Other needs**

871 Major needs in future in-transit UFP exposure studies have been outlined above, and  
872 numerous other aspects requiring additional research have been suggested throughout this  
873 review. Further investigation of the variability inherent in the determinants of exposure  
874 discussed in section 5 is required to permit better appreciation of their effects. There is also  
875 an obvious need for improved modelling techniques, incorporating GIS, and for further  
876 comprehensive assessments of the health risk-benefit balance for active transport modes (de  
877 Nazelle et al., 2009; de Hartog et al., 2010).

878

879 **11. CONCLUSIONS**

880 In our analysis of 47 studies comprising approximately 3000 trips undertaken in 6 transport  
881 modes, we found that highest trip-weighted mean concentration occurred in automobile  
882 cabins during tunnel travel ( $3.0 \times 10^5$  particles  $\text{cm}^{-3}$ ), and the lowest whilst cycling ( $3.4 \times 10^4$   
883 particles  $\text{cm}^{-3}$ ). Mean concentrations in bus, automobile (non-tunnel travel), rail, and walk  
884 modes were generally comparable. However, UFP exposure (and dose) during time spent in-  
885 transit is strongly dependent on a range of mode-specific and more general determinants,  
886 including, but not limited to, the effects of: meteorology, traffic parameters, cabin ventilation,  
887 filtration, deposition, UFP penetration, fuel type, exhaust treatment technologies, respiratory  
888 minute ventilation, route and microscale phenomena. Therefore, direct comparison of  
889 concentrations measured in different modes highlights general trends, but should not be  
890 extrapolated without detailed consideration of the above factors. Characterising the  
891 variability in the effects of these determinants will be an important aspect of future work.

892

893 There is preliminary evidence to suggest that time spent in-transit can contribute substantially  
894 to total daily exposure, and future studies require comprehensive assessment of 24 hour UFP

895 exposures across a broad demographic spectrum. Moreover, the range and variability of  
896 acute health effect associated with in-transit exposures are not well understood, and further  
897 studies are required to supplement the findings of the limited number performed to-date.

898

899 Transport is a ubiquitous component of life, and initial evidence suggests that UFP exposures  
900 incurred during this time can contribute substantially to daily exposure and be associated with  
901 adverse health effects in susceptible and healthy persons. Further research to better define  
902 this link is therefore well-justified, and will be of considerable benefit to urban planning,  
903 policy development and public health.

904

## 905 **ACKNOWLEDGEMENTS**

906 We thank Dr. Colin Solomon for his interest in this work and the useful feedback he  
907 provided.

908

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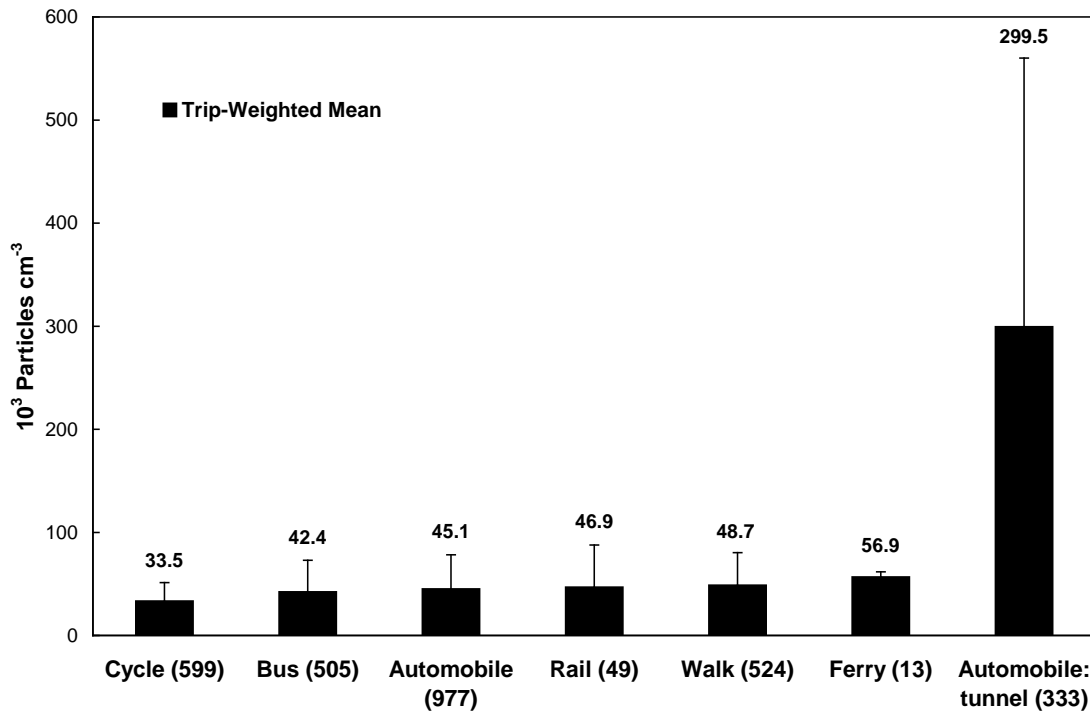
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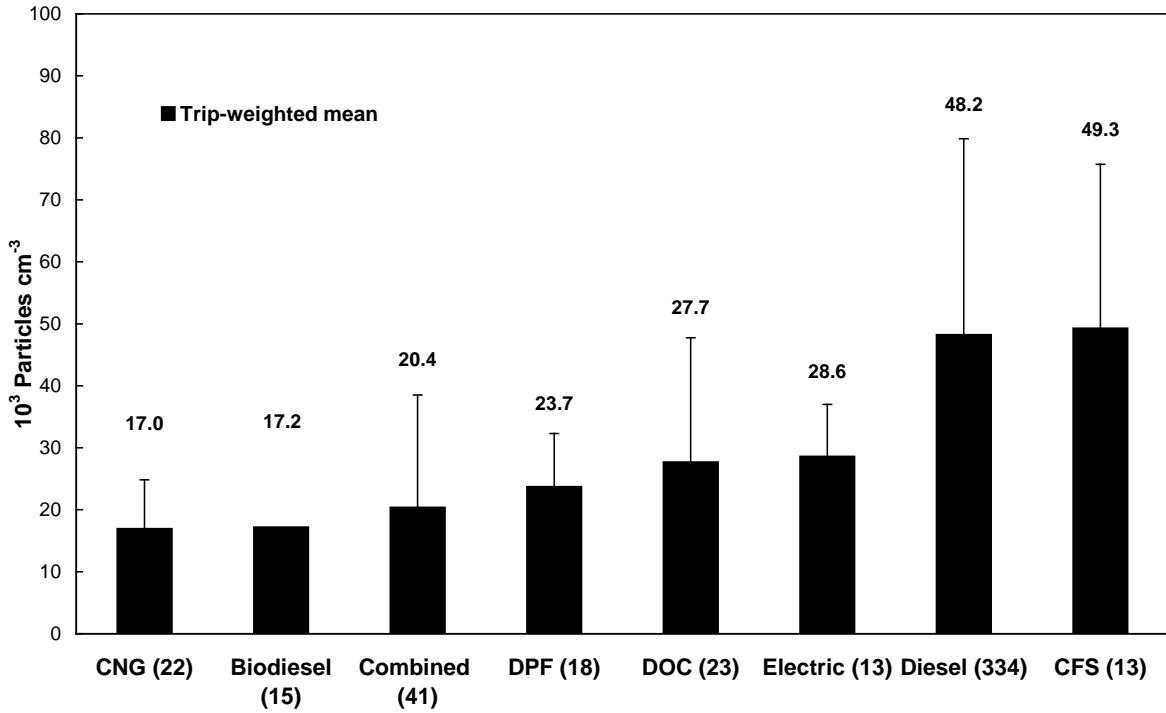
**Figure 1.** Trip-weighted mean UFP concentrations in each transport mode, shown as bold

1305 numbers. The number of trips taken in each mode is shown in brackets. Error bars denote

1306 the trip-weighted standard deviation. The studies from which the data were extracted are

1307 listed in the Supplementary Information file.

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1310 **Figure 2.** Trip-weighted mean UFP concentrations measured in buses of different fuel type  
 1311 and emission control device. The number of trips taken in each category is shown in  
 1312 brackets. Error bars denote the trip-weighted standard deviation. See text for abbreviations.

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