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**The Vehicle and the Environment:  
Urban Pollution Monitoring using both  
number- and mass-weighted particle  
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# **The Vehicle and the Environment: Urban Pollution Monitoring using both number- and mass-weighted particle measuring instrumentation.**

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Current ambient particle measurement strategies (for example  $PM_{10}$  and  $PM_{2.5}$ ) are mass-based and subsequently biased to the larger particles present in the environment (notably those associated with the classical accumulation and coarse particle modes). Since tail-pipe emissions from vehicles comprise significant particles in the ultra-fine region (condensation mode  $<0.1 \mu m$ ), which are not easily seen by the current mass-based measurement techniques, number-based techniques have been investigated as a more appropriate measurement of combustion sourced urban pollution. Detailed meteorological data coupled with traffic count data (by vehicle class and time) has been correlated with particle mass ( $PM_{10}$ ) and number concentration measurements over the size range 0.01 to  $10 \mu m$ . A clear correlation between traffic density and ultrafine particle concentration is observed whereas  $PM_{10}$  and traffic density show little correlation. Peak concentrations in excess of  $1 \times 10^6$  particles  $cm^{-3}$  were recorded indicating an increase of over 100x background.

Key Word Index: Traffic,  $PM_{10}$ , ultrafine, urban aerosol, pollution

## **INTRODUCTION**

Many studies have implicated particulate air pollution as contributing to the incidence and severity of respiratory disease (Dockery et al, 1989; Anderson et al, 1996.). More specifically, several recent studies have suggested an association between traffic-related air pollution and diminished pulmonary function and/or increased respiratory symptoms (Janssen et al. 1997, Weiland et al., 1994, Wjst et al., 1993).

Within the UK independent scientific committees reporting to Government have published reports indicating concern over the health effects of particle exposure. the Committee On Medical Effects of Air Pollution (COMEAP) and the Medical Aspects of Air Pollution Events (MAAPE). COMEAP conclude that it would be imprudent to ignore a probable causal link between particulate exposure and acute (and chronic) health effects. This is qualified, however, by the recognition that there is a degree of uncertainty over the role of very fine particles (i.e. those less than 0.1  $\mu\text{m}$  aerodynamic diameter). This uncertainty arises from the observation of a statistically significant increase in health effect markers resulting from small increases in  $\text{PM}_{10}$  concentrations (Lawther P J et al, 1968, King et al. 1997). The increase in observed mortality and morbidity is difficult to explain in terms of conventional toxicology and no plausible biological mechanism has been proven (Seaton et al., 1995). It was further noted, however that  $\text{PM}_{10}$  measurements could be insensitive to large increases in number concentration of very fine particles and that these smaller particles (generally from combustion sources) might offer a route where particle deposition in the lung manifests itself very differently from conventional inhalation toxicology (Seaton. (1996)).

On the basis of health effects data, the UK Expert Panel on Air Quality Standards (EPAQS) of the Department of the Environment has recommended that the 24 hour exposure limit be reduced from the current  $150 \mu\text{g m}^{-3}$  to  $50 \mu\text{g m}^{-3}$  (the current annual limit, as based on the current US EPA guidelines). In addition, the EPAQS recommends that efforts be made to further reduce the  $50 \mu\text{g m}^{-3}$  limit year on year, and that the number of days where the limit is exceeded is reduced year on year. However, the proposed UK National Air Quality Strategy indicates that to meet targets for PM exposure, it is expected that further control strategies will be required, including local air quality management, new vehicle and fuel standards, and 'a fierce crackdown on vehicle and industrial emissions'.

Beyond the UK, the European Commission is in the process of reviewing the health effects of particulate emissions. An Air Quality Framework Directive has been adopted with a position paper and draft daughter Directive focused on Particulate Material expected during 1997. This is expected to be similar to UK and US approaches in reducing 24 hour  $\text{PM}_{10}$  limits to  $50 \mu\text{g m}^{-3}$  with limits on permitted exceedances. In conclusion, there is a body of evidence to suggest that future emissions legislation within Europe and the US will include a reduction in particulate emission levels, coupled with a degree of size discrimination. In Europe, number concentration limits may also be adopted depending on the outcome of new research programmes.

Current emissions legislation (for example, Directives 70/220/EEC and 96/69/EC - Measurement of Light Duty Emissions) is based on the total mass of particles emitted per km, with environmental legislation based on a mass per unit volume basis with

no reference to the size of the particles or the number concentration of particles emitted. There is, however, a growing body of concern over the health effects of inhaled particles from combustion sources and the potential influence of both the chemical composition and the size distribution of the emitted particles (Dockery et al., 1993, Rickeard et al., 1996, Seaton 1995, 1996 Stern et al., 1973).

Furthermore, a recent study, based on  $PM_{10}$  measurements, examining the micro- and macro-control options available for implementing better local air quality management (for example, an integrated environment and traffic policy) suggested that  $PM_{10}$  levels were largely unaffected by local emissions and were the result of macro-effects (Stedman 1997). For example, large scale movement of particulates over the UK and in some cases over continental Europe.

This observation and/or conclusion raises the issue of whether or not  $PM_{10}$  can be used as firstly, a local pollutant control indicator for traffic or industry and secondly, whether it is the most appropriate measurement of Urban pollution and in particular when traffic pollution is considered to be a major source.

With the increasing public concern over the potential health-related issues associated with ultra- fine particles (for example  $<100$  nm), this study was directed at examining the link between "tail-pipe" emissions (measured on a light-duty dilution tunnel) and ambient particulate levels (measured in urban locations). Initial studies indicate that the majority of the vehicle-produced particles in the ambient environment are less than 100 nm. Although these exist at very high number concentrations (for example observed peaks  $> 10^7$  particles  $cm^{-3}$  and hourly time-averaged concentrations  $> 4 \times 10^5$  particles  $cm^{-3}$ ) they have very little mass associated with them ( $<10 \mu g m^{-3}$ ). A recent study in the centre of Oxford, a medium-sized UK city using state-of-the-art aerosol research equipment illustrates these issues.

## METHODS

### *Study Description*

Over the Easter Bank Holiday weekend (28 March to 2 April 1997), a particulate monitoring station was located in the centre of Oxford (Carfax Tower, Grid Ref. SP513062, Figure 1) an inland city in the UK with approximately 135,000 inhabitants and an economic mix of tourism, academia and light-to-medium industry. The city centre location was chosen since it was near (10m) a busy cross-roads (typically 8000 vehicles per day), but more importantly, because it was close to the shopping areas(s) and the city centre bus drop-off and pick-up points.

The kerbside particulate monitoring station housed a PM<sub>10</sub> monitor (TEOM Series 1400 : Rupprecht & Patashnick (1991)), a TSI Inc. Aerodynamic Particle Sizer (APS : Hering S V (1989)), a Climet Optical Particle Counter (OPC : Hering S V (1989)) and a TSI Inc. Condensation Nucleus Counter (CNC : Hering S V (1989)). This suite of instruments enabled real-time measurements of PM<sub>10</sub> mass concentrations, and particle number-concentrations over the size ranges >0.01, >0.5 and 0.5<Dp>10 µm for the CNC, OPC and APS respectively.

Data from a second PM<sub>10</sub> monitor (TEOM model 1400a) located to the North of the city was provided by Oxford City Council (Grid ref. SP518066) approximately 800 m from the congested city centre.

**TEOM:** The Rupprecht and Patashnick Tapered Element Oscillating Microbalance (R. & P. TEOM) instrument offers filter-based real-time monitoring of gaseous particulates (Rupprecht and Patashnick (1991), Patashnick and Rupprecht (1986) and Patashnick and Rupprecht (1991)). The tapered element assembly is vibrated at its natural frequency to provide a real-time mass response by assuming that it behaves as an undamped oscillator. The sample is passed through the filter at a controlled and measured flow rate, thus providing particulate concentration information.

The sensor is calibrated by measuring the periods with,  $\tau_1$ , and without,  $\tau_2$ , a known mass (pre-weighted filter cartridge):

$$k_0 = \frac{dm}{(\tau_1^2 - \tau_0^2)}$$

The sampling station contained a Model 1400 TEOM system fitted with a Graseby Andersen PM<sub>10</sub> sampling head (Model P29). The unit / software was configured to meet the US EPA equivalence criteria for PM<sub>10</sub> monitoring.

**APS:** The aerodynamic particle sizer (Model APS33B, TSI Inc., St. Paul, MN) is a real-time instrument for determining the aerodynamic size of aerosol particles ranging from 0.5 to 30 µm aerodynamic diameter (limited in this study to 10 µm by the

PM<sub>10</sub> sampling head); the aerodynamic diameter is defined as the diameter of the unit density sphere having the same settling velocity as the particle under consideration. The instrument operates by time-of-flight measurement of individual particles passing through a split laser beam under an accelerating flow field. The recorded time-of-flights can be related to the aerodynamic sizes of the specified particles using an instrument-specific calibration curve (Remiarz et al., 1983).

**OPC:** The optical particle counter (Climet ) measures the quantity of light scattered by individual particles as they pass through a beam of intense light. The amount of scattered light is measured by a photosensitive detector. The magnitude of the electrical signal is proportional to the number of particles.

**CNC:** The sampling station was equipped with a condensation nucleus counter (Model 3020, TSI Inc., CNC). This instrument is capable of measuring the number concentrations of particles >10 nm up to a concentration of 10<sup>7</sup> particles cm<sup>-3</sup>. Particles entering the instrument pass through a vapour source, in this case butanol, which subsequently condenses on the particle. The resulting increase in particle size enables the particles to be countered by measuring the scattered light as in a traditional OPC. At high concentrations >10<sup>3</sup> particles cm<sup>-3</sup> significant coincidence problems prevent single count mode operation and the instrument switches into a photometric mode. In this mode, the photodetector voltage is converted to a concentration by means of an internal calibration. The continuous analogue outputs from the CNC were directly recorded at a sampling rate of 1 per second through a 16 bit A/D card coupled with a Labview software package onto a desktop PC. This fine time resolution coupled with the high instrument sensitivity (1 to 1x10<sup>6</sup> #cm<sup>-3</sup>) enabled the data to be examined on both a short- (discrete data) and long-time basis (hourly averaged values).

**Golden River Traffic Classifier:** Inbound and outbound hourly traffic counting was carried out at St. Aldates (Grid Ref. SP510060) by using an automatic traffic counter and classifier. This instrument was capable of classifying the vehicles into 6 classes (Table 1) by using inductive loops set into the road surface.

**Table 1. Vehicle Class description**

CLASS	DESCRIPTION	LENGTH RANGE (M)	CHASSIS LEVEL
1	Motorbike	0.00 - 2.50	X
2	Car/Van	2.50 - 4.80	X

3	Car + Trailer	7.00 - 10.00	Low
4	LGV	4.80-6.00 / 6.00-7.00	X / Low
4	Rigid HGV	6.00 - 11.00	High
5	HGV	11.00 - 18.00	High
6	Bus	10.00 - 18.00	Low

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Hourly meteorological data (temperature, wind speed, wind direction, humidity, sunshine hours and precipitation were supplied by the UK Meteorological Office for Brize Norton (22 km West of Oxford at Grid Ref. SP290060) and wind for Oxford city centre by OCC.

## RESULTS

### Traffic Data

Traffic data for the 6-day sampling campaign was recorded on an hourly basis. The sampling period contained 2 normal working days (Thursday 27/03/97 and Tuesday 01/04/97), 2 public holidays (Good Friday and Easter Monday) and a Saturday and Sunday. Surprisingly, the recorded daily traffic flows were largely invariant to the day of the week (Fig 1a) and the total daily traffic numbers recorded were typically 8,000 vehicles per day (Fig 1b). This probably reflects Oxford being a significant tourist centre. However, the class breakdown of the total traffic numbers shows a clear correlation to public transport policy on Bank Holidays and Sundays (Fig 1c). On these days the bus numbers were significantly smaller (102, 58 and 65 for Friday, Sunday and Monday respectively) which compared with approximately 500 on a normal work day.

### Meteorological Data

During the 6-day sampling period the weather was dry, reasonably calm, sunny and warm. Wind speed measurements at both Brize Norton and Oxford showed good general agreement (Fig 2a). However the first 2 days had considerably windier conditions than recorded in the subsequent 4 sampling days (24 and 18 knots for Thursday and Friday respectively compared with <8 knots

for the remaining period). This small change in wind speed from a meteorological perspective has dramatic effects on pollutant dispersion as will be shown below.

The temperature (Fig 2b) and humidity (Fig 2c) profiles show large daily swings consistent with sunny conditions (7-11 sunshine hours per day recorded). Throughout the sampling period, the wind direction remained approximately south-westerly (Fig 2d). In conclusion, the weather conditions over the 6-day sampling period were ideal for a short-term ambient particulate sampling campaign. Except for wind velocity, which initially peaked at approximately 20 knots for the first two days before becoming calm for the remaining time (<8 knots). Wind direction, humidity, temperature profile and daily sunshine hours were reasonably constant.

### **PM<sub>10</sub> Data**

Hourly averaged PM<sub>10</sub> particle concentrations were recorded throughout the sampling campaign at both the curb-side mobile laboratory at Carfax (AEA) and on the roof of the Oxford University School of Geography (OCC). The latter instrument is managed and controlled by Oxford City Council (OCC); and provides their continuous ambient particulate monitoring. The data from these two instruments are given in Fig 3, and Table 2 with some additional data from the OCC instrument.

Fig 3 demonstrates that the recorded particulate mass at both locations appeared very similar and throughout the sampling campaign the hourly averaged concentrations were less than the 50  $\mu\text{g m}^{-3}$ . Correlating the two hourly averaged instrument responses indicates agreement within  $\pm 10 \mu\text{g m}^{-3}$  (95% confidence interval), with a gradient of 0.988 (linear regression with intercept set to zero) and a  $r^2 = 0.602$ . The daily averaged variation between the two instruments is, however, much smaller ( $< 2 \mu\text{g m}^{-3}$ ) and well within the instrument measurement errors (Table 2). The reason why the hourly averaged data is more scattered, albeit with a small absolute variation, can be attributed to both the fundamental poorer instrument resolution over the shorter averaging period and to the reduced TEOM performance caused by the less stable measurement platform (for example errors introduced via instrument vibration) in the mobile laboratory. It should be noted that no systematic differences between the two instruments were observed as a function of date or time.

**Table 2. Daily averaged PM<sub>10</sub> values**



	27/03/97	28/03/97	29/03/97	30/03/97	31/03/97	01/04/97
AEA PM <sub>10</sub>	-	10.0	16.7	14.3	16.4	-
OCC PM <sub>10</sub>	16.7	11.8	14.2	14.8	17.6	24.7

During the 6-day sampling period the range of 24 hr averaged PM<sub>10</sub> was 12 and 25  $\mu\text{gm}^{-3}$  and therefore under the draft UK National Air Quality Strategy the particulate air quality was classified as very clean.

Without reference to the other instrument data, these results on their own clearly indicate poor instrument sensitivity to tail-pipe emissions, when integrated with the full spectrum of particles associated with ambient PM<sub>10</sub> (combustion, accumulation and weathering particulate) at low absolute level, since there is little doubt that the concentrations of tail-pipe particles emitted in the vicinity of the curb-side monitor are significantly greater than at the OCC monitor. the performance of the TEOM for undertaking quantitative measurements of tail-pipe generated particles is not in question since several laboratory-based studies have clearly shown good correlation's with regulated-filter mass concentration measurements. (Baron 1988, Allen, G et al 1997) but at higher absolute mass levels. However, this limited data set suggests that the short-term tail-pipe contribution to PM<sub>10</sub> at the Carfax monitoring site is lower than expected and probably less than 5  $\mu\text{gm}^{-3}$ .

Although this observation appears to contradict current PM<sub>10</sub> source term apportionment estimates (for example vehicles contribute 26 % (DETR, 1997)) this is not necessarily the case. For example, tail-pipe-based emissions in the environment are likely to comprise both "young" (particles directly leaving the tail-pipe) and "old" (particles subject to environmental based changes, for example, photo-chemistry) particles that are not necessarily the same in either concentration or size. Thus, the overall contribution from the tail-pipe to ambient PM<sub>10</sub> must examine both these potential contributions. This point is re-addressed later in this paper.

### APS Data

The APS measures the number-weighted aerodynamic size distribution of aerosol particles ranging from 0.5 to 10  $\mu\text{m}$  aerodynamic diameter. Throughout the sampling campaign, RS232 communication problems were encountered which resulted in an incomplete data set. The instrument was set to record on a 5 minute interval. Typically, the number-weighted medium diameter was centred at around 1  $\mu\text{m}$  aerodynamic diameter (Fig 4) consistent with the accumulation mode, with typical corresponding number concentrations of less than 5 particles  $\text{cm}^{-3}$ .

By assuming that the particles had both a mean particle density of  $1 \text{ g cm}^{-3}$  and were spherical (as would be expected from the accumulation mode), a corresponding mass concentration can be calculated from the discrete size channel data. These data are shown in Fig 4c. Correlating these data with the TEOM measured mass concentrations indicates good overall agreement;  $3 \pm 6 \text{ } \mu\text{gm}^{-3}$  (95% confidence interval). Thus, it appears that much of the ambient  $\text{PM}_{10}$  mass comprises particles in the  $0.5$  to  $2 \text{ } \mu\text{m}$  aerodynamic size range and consequently supports the low relative and absolute contribution of “young” tail-pipe particulate to ambient  $\text{PM}_{10}$  (see  $\text{PM}_{10}$  measurements above). No significant numbers of coarse particles (i.e. particles  $> 3 \text{ } \mu\text{m}$  aerodynamic diameter) were evident, and thus the traditional coarse particle mode (particles associated with mechanical generation, for example, wind blown dust, tyre dust, brake dust) were not present during the study. This may simply reflect of the relative calm weather throughout the sampling campaign and thus low levels of re-suspension.

### OPC Results

Particle counts for all particles greater than  $0.3 \text{ } \mu\text{m}$  were measured using the OPC. These are shown in Fig 5 and are consistent with the particle number data from the APS, as would be expected from their respective measurement ranges.

### CNC Results

The hourly time-averaged particle number data are given in Fig 6a for the 6 day sampling period. These data indicate peak daily concentrations in the range of  $10^5 \text{ particles cm}^{-3}$  and, moreover, indicate that the daily shape (although not necessarily the magnitude) of the ultrafine number concentrations are very similar; concentrations start rising at about 0800 hrs GMT, peak around 1300 hrs and fall to near background levels at 1800 hrs. This is contrary to observations with the OPC, APS or TEOM where no clear daily profiles were evident. Although the traffic count showed similar daily profiles throughout the study, the particulate concentrations in the first two days were considerably lower than in the subsequent days. This however, is entirely consistent with the recorded wind speeds (i.e. significantly higher on the first two days) and therefore likely to be simply a function of increased wind-related dispersion. The peak concentrations on the Sunday and Monday were similar,  $2 \times 10^5$  and  $3 \times 10^5 \text{ particles.cm}^{-3}$  respectively, and reflect similar weather and vehicle flows. The peak concentration on the Tuesday is greater than that observed on the Sunday or Monday and, again, is consistent with an increase in business traffic (for example buses, LGVs etc.). Background levels (i.e. those measured during periods of low traffic flows (e.g. 0300 - 0500 hrs)) were consistent throughout the sampling campaign (i.e.  $1 \times 10^4$  to  $3 \times 10^4 \text{ particles.cm}^{-3}$ ).

Fig 6b demonstrates that, the raw instrument data (1 s time base) exhibits large short-term fluctuations. For example, the range of concentrations recorded between 1200 and 1205 on the 4th day was  $4 \times 10^4$  to  $4 \times 10^5$  particles  $\text{cm}^{-3}$ . This is likely to be associated with single events such as a vehicle passing the sampler.

Although consistent background levels for all instruments were recorded daily between 0300 and 0500 Hrs GMT in Central Oxford, it was decided to measure rural background levels away from significant traffic flows. The mobile laboratory was removed at 1200 Hrs on the Tuesday and driven 10 miles south-south-west from Oxford to a small village. At this location, the CNC recorded a relatively low and reasonably constant ultrafine particulate levels of  $1 \times 10^4$  particles. $\text{cm}^{-3}$  for 24 hrs (Fig 6d).

A parallel study (unpublished data) demonstrates sensitivity of this CNC technique to monitoring traffic tail-pipe emissions; a UPM was located next to a quiet rural road (typically less than 20 vehicles per hour) with a video camera. Each vehicle passing the sampler resulted in a clear particulate emission peak (Fig 6c). (The arrows on Fig 6c represent each vehicle movement. The delay in instrument response is simply the time taken for the particles to reach the instrument inlet plus the instrument measurement time (estimated to be in total about 20 seconds). Although the magnitude of the peak varied from one vehicle to another, peaks from both diesel and gasoline cars were always observed and moreover were several times the ambient background levels.

### **Instrument Comparison**

The hourly averaged data from each of the 4 instruments located in the mobile laboratory have been normalised to 0300 GMT. on the second day to enable their respective relative changes to be studied (Fig 7). The relative changes during daily cycles for the CNC are significantly higher (relative change  $>20$ ) than those recorded with the other 3 instruments (relative change  $< 2$ ). Moreover, on closer inspection it can be seen that daily profiles measured by the CNC are both reproducible and in phase with city activity and in particular vehicle movements. The higher mean wind speeds recorded in the first few days, resulting in greater dispersion / dilution, are likely to be the major cause of the reduced particulate number levels recorded.

## Traffic Correlation

The correlation between the inbound traffic flow and the CNC hourly averaged data is shown in Fig 8. The data clearly show a good overall correlation with much of the fine detail showing on both traces. For example, the shoulder on the main traffic peak measured during the 30<sup>th</sup> and 31<sup>st</sup> March is clearly matched on the hourly averaged CNC trace. Although the traffic profile is broader than the pollution profile, this can be explained in terms of congestion. During the early hours and late evening, the traffic moves at a greater average speed than during the normal "business hours".

## Conclusions

This study supports the hypothesis that in many urban locations, PM<sub>10</sub> is not the optimum local pollution monitoring parameter. Much of urban PM<sub>10</sub> is simply macro-scale movements of the secondary accumulation mode aerosol and thus difficult to manage in a local micro-environment. This view is supported by the work of Stedman, 1997.

Although these high number-based levels may not be significant from a traditional mass-based health perspective, recent epidemiological studies have, albeit not conclusively, shown a link between ultrafine particles and diminished pulmonary function and/or increased respiratory symptoms in people (Janssen et al 1997, Weiland et al., 1994, Wjst et al., 1993).

The very high numbers of particles being emitted from vehicle tail-pipes (for example  $>1 \times 10^{10} \text{ s}^{-1}$ ) enable vehicle specific pollution monitoring to be carried out easily.

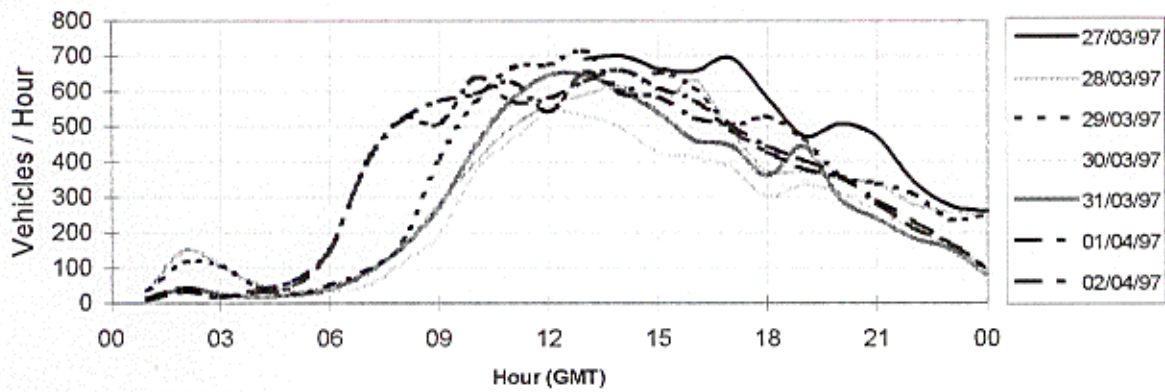
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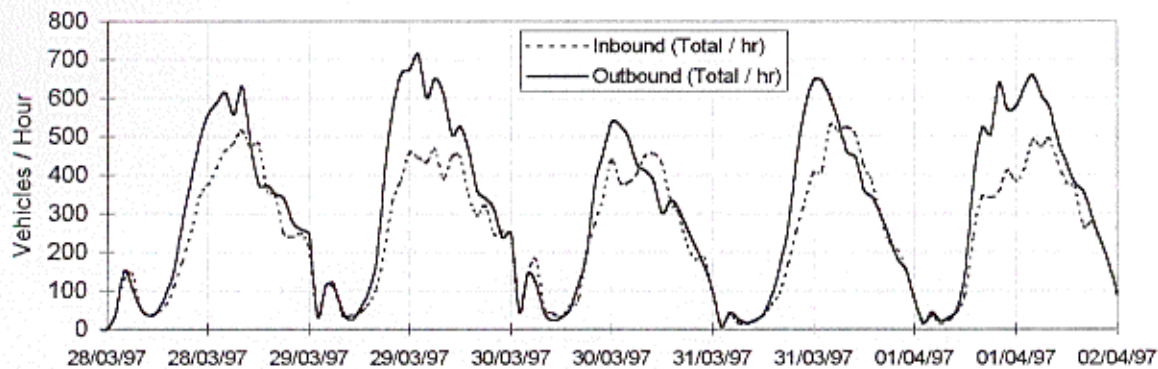
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**Figure 1. Traffic Data**

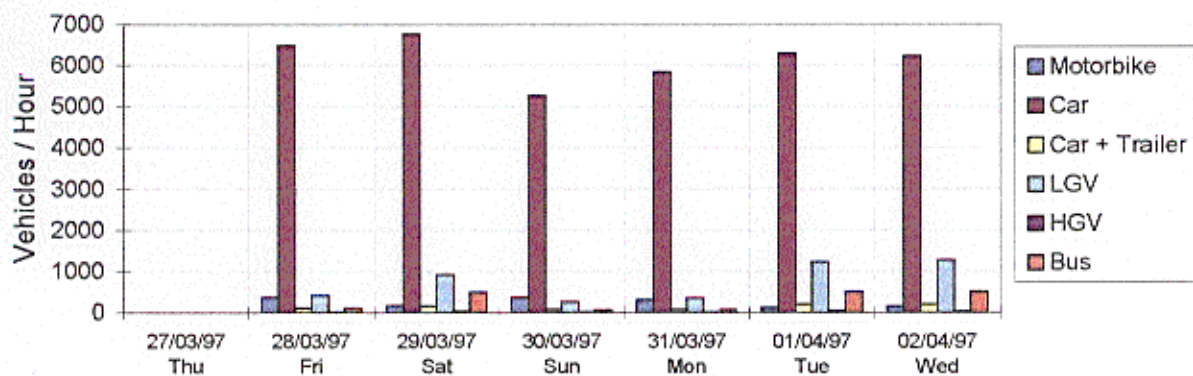
**a) Daily Vehicle Profile**



**b) Inbound and Outbound Daily Profiles**

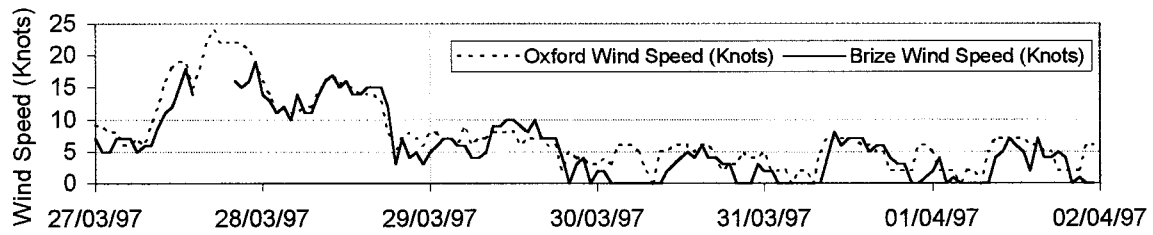


**c) Class Profile (Outbound)**

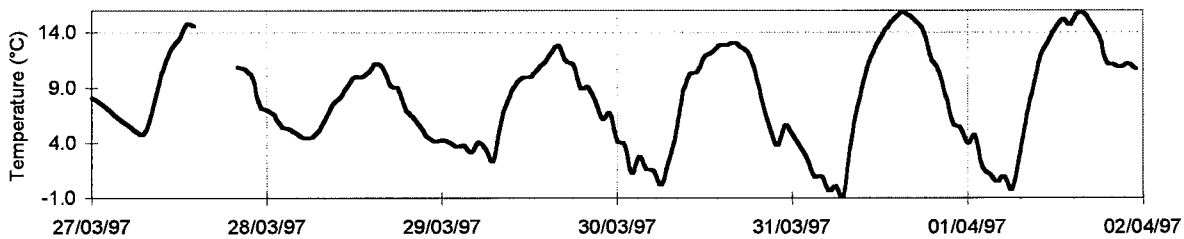


**Figure 2. Meteorological Data**

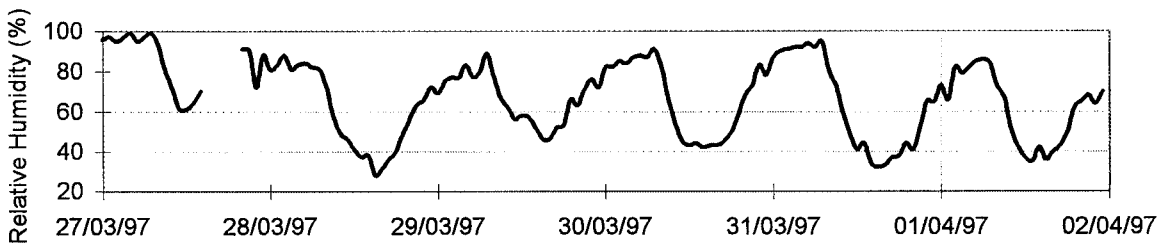
**a) Wind Speed**



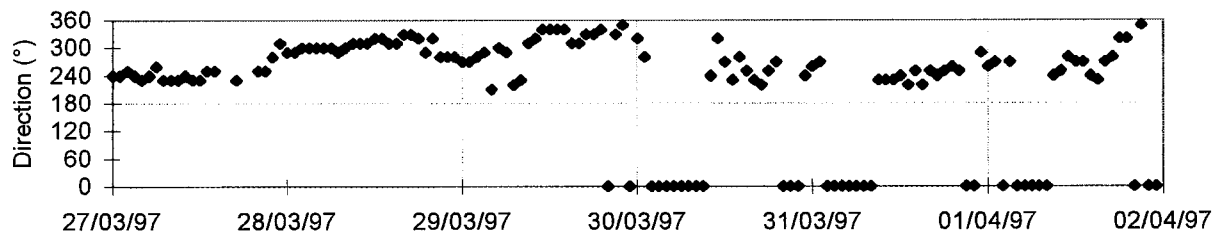
**b) Temperature**



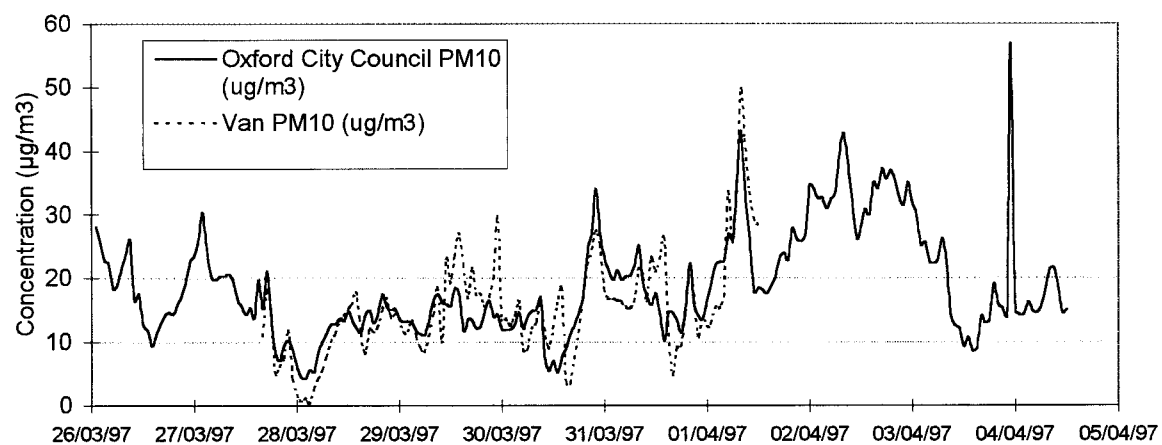
**c) Humidity**



**d) Wind Direction**



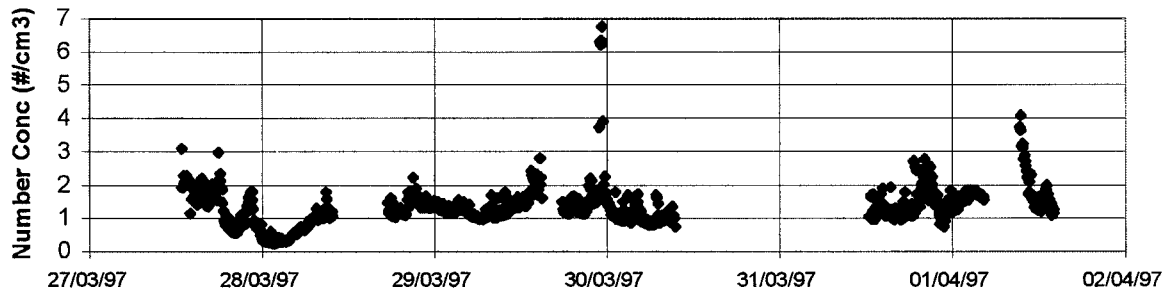
**Figure 3. PM10 Results at both Carfax and at the School of Geography**



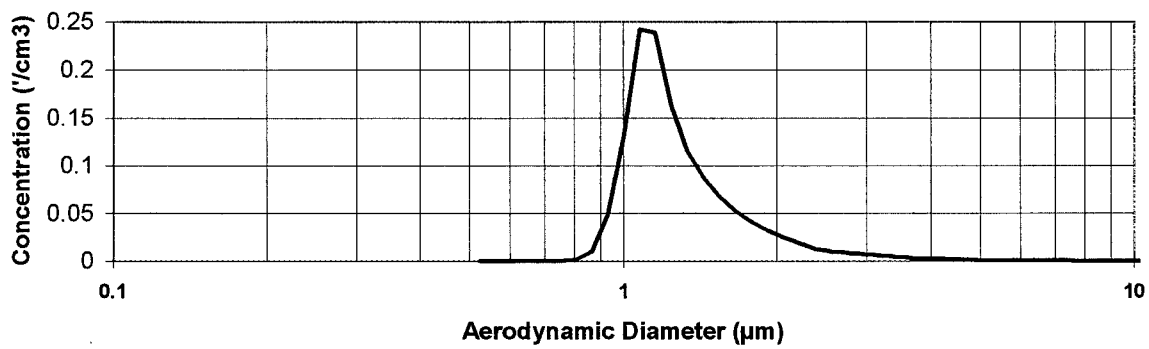


**Figure 4. APS Data**

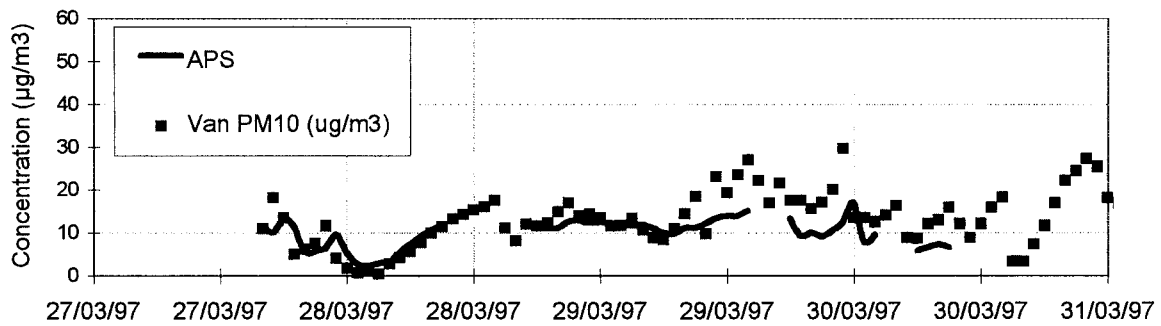
**a) Number Concentrations**



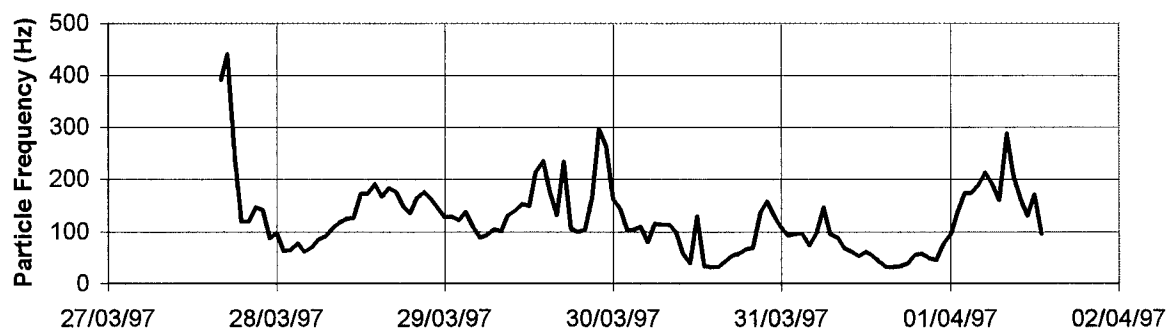
**b) Number-weighted aerodynamic particle size distribution (average)**



**c) Mass-weighted Concentrations**

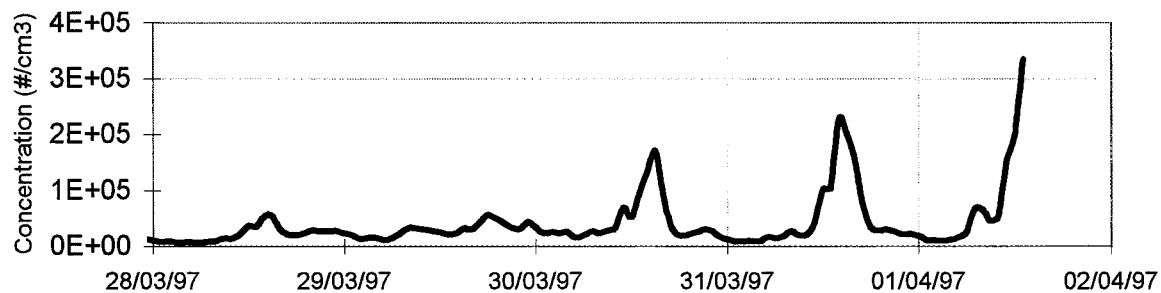


**Figure 5. OPC Results**

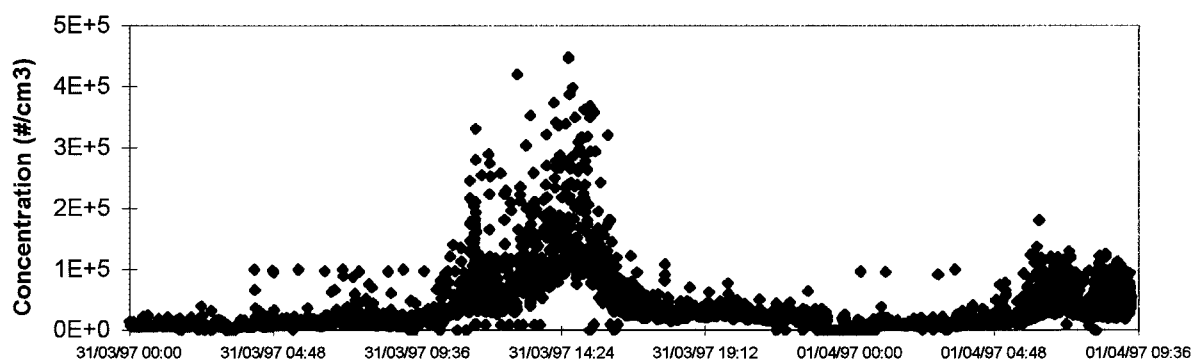


**Figure 6. CNC Results**

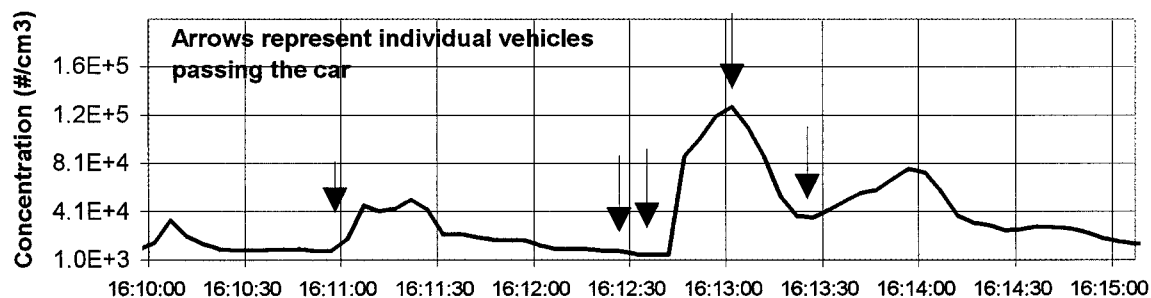
**a) Hourly averaged data**



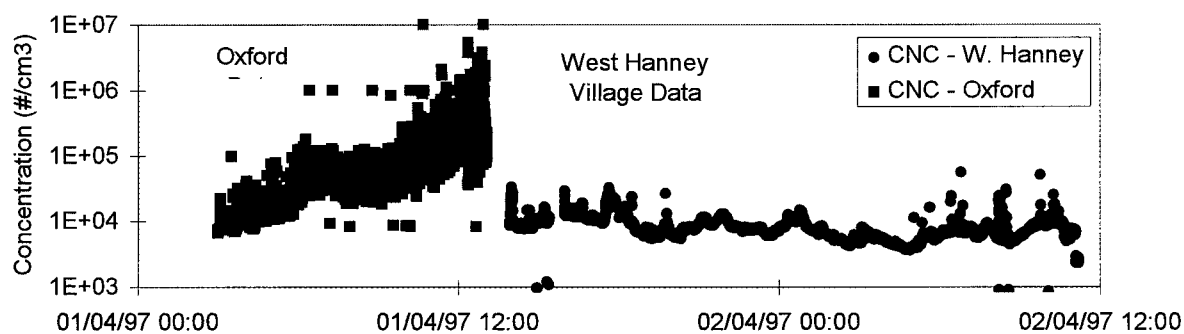
**b) Raw Data**



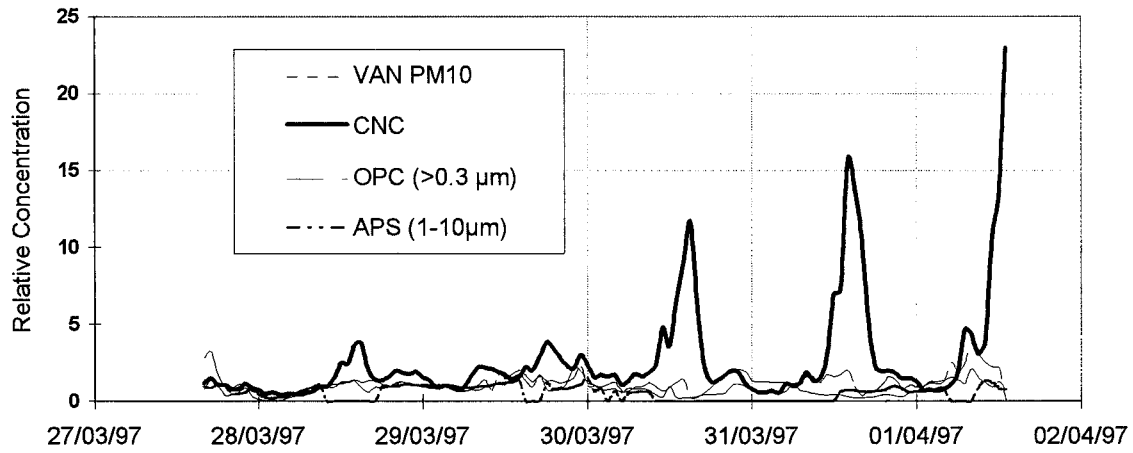
**c) Individual Vehicle**



**d) Rural Comparison**



**Figure 7. Instrument Inter-comparison**



**Figure 8. Traffic Correlation**

