COVER SHEET

Morawska, L and Hofmann, W and Hitchins-Loveday, J and Swanson, C and Mengersen, K (2005) Experimental study of the deposition of combustion aerosols in the human respiratory tract. Journal of Aerosol Science 36:pp. 939-957.

Experimental Study of the Deposition of Combustion Aerosols in the Human Respiratory Tract.

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Abstract

The total deposition of environmental tobacco smoke (ETS), diesel and petrol smoke in the respiratory tract of 14 non-smokers between the ages of 20 and 30 was determined experimentally. A Scanning Mobility Particle Sizer (SMPS) measuring a size range of 0.016 – 0.626 µm was used to characterise the inhaled and exhaled aerosol during relaxed nasal breathing over a period of 10 minutes. The ETS, diesel and petrol particles had average count median diameter (and geometric standard deviation) of 0.183 µm (1.7), 0.125 µm (1.7) and 0.069 µm (1.7), respectively. The average total number deposition of ETS was 36% (standard deviation 10%), of diesel smoke 30% (standard deviation 9%), and of petrol smoke, 41% (standard deviation 8%). The analysis of the deposition patterns as a function of particle size for the three aerosols in each individual showed that there is a significant difference between each aerosol for a majority of individuals (12 out of the 14). This is an important result as it indicates that differences persist regardless of inter-subject variability.

Introduction

Knowledge of the deposition of particulate matter in the human respiratory system is important for dose assessment and the risk analysis of airborne pollutants. Deposition process is controlled by physical characteristics of the inhaled particles and by the physiological factors of the individuals. Of the physical factors, particle size and size distribution are among the most important ones. Over the recent years an increasing attention has been devoted to the particles in the smaller end of the size distribution, which are submicrometer and ultrafine (< 0.1 μm) ranges. This is because on the one hand these particles can penetrate to the deeper parts of the respiratory tract, and on the other hand, they are generated in abundance by the most significant pollution sources, which are those related to combustion processes. In addition, it has been reported that ultrafine particles show exceptional toxicity with regards to lung morbidity and mortality (Donaldson *et al.*, 1998); (Oberdorster *et al.*, 1995). Most of the particles generated from combustion processes are in the submicrometer range, usually around 0.01 – 0.3 μm. They have an almost insignificant contribution to the mass of airborne particulate matter currently used in air quality standards; (eg. PM_{10} and $PM_{2.5}$, the standard measures of the mass of particulate matter less than 10 μm and 2.5 μm, respectively). However, in terms of number, these particles constitute the vast majority of all the airborne particles, usually over 90% and above. Thus knowledge of the lung deposition of combustion aerosols and more generally, aerosols in urban air, is important because of this combination of the particle abundance and toxicity.

Whilst a number of theoretical modelling studies of lung deposition have been performed (presented in (Stahlhofen *et al.*, 1989)), the majority of them focused on particles >0.1 μm, and are supported by experimental studies in this range. Relatively few experimental data exist for particles <0.1 μm, and only one study (, included a larger group of subjects of 22, as opposed to 5 or less for other studies, as summarised in the paper).

In particular, (Stahlhofen *et al.*, 1989) presents a model of total deposition versus diameter (from 0.005 – 10 μm) using experimental results obtained by two other authors. However, the diameter of the particles is for that of unit density spheres and as such, is not directly applicable to the particles from combustion sources, which are much more complex in terms of their morphology. (Jaques and Kim, 2000) generated aerosols (non-hygroscopic metallic nuclei particles coated with sebacate oil) of number median diameter 0.04, 0.06, 0.08, and 0.1 μm, (geometric standard deviations - GSDs of 1.3) and examined the total deposition at 10 lung depths ranging from 50 – 500 ml. The focus of the study was on the effects on total deposition of different breathing patterns and of gender, over four size distribution ranges. The authors found that the deposition of ultrafine particles increases with the decreases of particle size and with breathing patterns of longer respiratory time. They also showed that there was a differential lung dose of ultrafine particles and thus there may be a differential health risk for men versus women. However, as only four size distributions were generated, the fractional deposition could not have been examined in detail with a view toward application of the results for modelling. A study of the literature by (Morawska *et al.*, 1999) reported that lung deposition varies widely between experimental studies, and that many theoretical models predict lower depositions than the experimental values. This is probably due to a number of factors including the differences in types of aerosols used, size ranges, breathing patterns, age and number of subjects, and measuring equipment.

The deficiencies in the experimental database and the discrepancies between the studies described above make it difficult to validate the existing theoretical models. Therefore the objective of this study was to experimentally investigate the total and fractional deposition in the human respiratory system of three common types of combustion aerosols, including environmental tobacco smoke (ETS), diesel and petrol emissions from motor vehicles. These aerosols are significant and prolific air pollutants to which a large majority of people are exposed.

This study is a continuation and expansion of the previous study conducted by the authors (Morawska *et al.*, 1999), (Hofmann *et al.*, 2001), which showed that the total particle number concentration of ETS deposited in the human respiratory system was about three times higher than that predicted by theoretical models. The present study aimed to expand the previous experimental investigations by also including aerosols other than ETS (diesel and petrol emissions). It also aimed at testing and interpreting the findings against current theoretical models of lung deposition. The focus of this paper is on the former (experimental study), while the latter (modelling) is presented in a companion paper.

Methods and Techniques

In this study, the number concentration and size distribution of combustion aerosol particles in the submicrometer range inhaled and exhaled by 14 human volunteers was determined using a Scanning Mobility Particle Sizer (SMPS). The subjects were in a relaxed state and inhaled nasally using a spontaneous breathing pattern. The test aerosol concentrations were well below high concentrations commonly encountered near the emission sources, such as busy road intersections (petrol and diesel particles) or public bars (ETS). The measurements were carried out in the International Laboratory for Air Quality and Health (ILAQH) at the Queensland University of Technology (QUT). The subjects were examined by qualified medical personnel at the QUT Health Services Centre where their lung capacity and normal breathing rate were determined.

Subjects

Selection process and criteria

The participants of the study were to be non-smokers and non-asthmatics of either sex and to be between the ages of 20 and 30. It was considered important to restrict the age of participants to a relatively narrow age band in order to minimize the effect of age as a possible factor affecting lung deposition. This particular age band was chosen to be the same as in the previous study conducted by the authors (Morawska *et al.*, 1999). In order to recruit participants for the study an email was sent to all the staff and students at QUT requesting volunteers for the project. The total number of participants recruited for the study was 14, and was sufficient to ensure statistical power of the study, as explained below.

Statistical determination of the number of subjects required

It was necessary to determine the number of people to be tested in order to conclude that a difference between theoretical and experimental nasal deposition rates existed and that this difference was of sufficient size to have occurred by some means other than chance. The experimental nasal deposition rates from the (Morawska *et al.*, 1999) study and the theoretical nasal deposition rates from (Hofmann *et al.*, 2001) were used in the following calculations.

Calculating the sample size:

1. “Practical important difference”: δ = deposition(theor) – deposition(exp)
2. “Combined standard deviation”: s = SQRT[(variance(theor) + variance(exp)]
   { Standard deviation = SQRT(variance). The variance of a difference is the sum of the two variances.}
3. “Significance level”: α (to control the probability of claiming a significant difference when there isn’t one, usually set to be .05)
4. “Power”: (1-β) (to control the probability of claiming no significant difference exists when there really is one, usually set to be .20)
5. Sample size “n”:

   If test is significant:

$$\frac{\delta}{s/\sqrt{n}} > \left(z_{\frac{\alpha}{2}} + z_{1-\beta} \right)$$

$$\text{This gives : } \quad n > \left((z_{\frac{\alpha}{2}} + z_{1-\beta}) \times {}^{s}\!/_{\delta} \right)^2$$

   where, z is the critical value, that is the positive z value that is at the vertical boundary for the area of $\alpha/2$ or (1-β) in the right tail of the standard normal distribution.

In this case for nasal deposition:

Dep (theor) = 16.9% ± (2.2 SD) i.e. variance(theor) = 4.84

Dep (exp) = 56% (±15.9 SD) i.e. variance(exp) = 252.81

Given that the intra-subject variability was ≈ 12% (Morawska *et al.*, 1999), a practical important difference would require to be 15%:

$$\delta = Dep_{theor} - Dep_{\exp} = 15\%$$

$$s = \sqrt{4.84 + 252.81} \approx 16$$

$$z_{\frac{\alpha}{2}} = 1.96 \qquad (\alpha = 0.05)$$

$$z_{1-\beta} = 1.28 \qquad (1-\beta = 80\%)$$

$\therefore$n > (3.24 x 16/15)^2

n > 11.9

This means for a sample size of 12 people or greater, a 15% or greater difference between the average theoretical and experimental nasal deposition represents a difference that would occur due to chance in only 5% of such experiments.

Equipment

Instrumentation

The submicrometre particles were characterised using the TSI (TSI Incorporated, St. Paul, MN, USA) Scanning Mobility Particle Sizer (SMPS) (Model 3071) consisting of an Electrostatic Classifier (EC) and a Condensation Particle Counter (CPC Models 3022A and 3010 were both used over the study period). An impaction nozzle (at the inlet of the EC) of diameter 0.0508 cm, and an aerosol flow rate of 0.3 L.min^{-1} were used to give a measured size range of 0.016 – 0.626 µm. (Note that the 3022A model was used for 9 subjects with a range of 0.016 – 0.626 µm, whilst the 3010 model was used for 5 subjects and gave a range of 0.015 – 0.670 µm.) The time taken to measure one sample of aerosol was 90 s (60 s of measurement with a 30 s delay between samples).

Set-up

The experimental set up for this study was based on the previous study by (Morawska *et al.*, 1999). Two aerosol chambers of volumes of 1 and 3 m^3 were used in the study, one in order to allow the deposition measurement of three aerosols for each volunteer in a single session to be conducted. Since some time is needed to flush a chamber and re-introduce another aerosol to the required concentration, availability of two chambers for the measurements enabled using the second chamber, while the first was flushed to remove the first aerosol, which was inhaled by a volunteer. The set-up is shown in Figure 1. Samples of the source aerosol were taken from points “1” on the relevant chamber and from point “2” on the sampling box. The vent in the 9 L sampling box was always open to allow pressure equalisation whilst the subject was breathing. The HEPA filter and pump were used to flush out the sample chamber between different aerosol tests. The mask was a standard air-purifying respirator, modified such that the subject inhaled aerosol from the environmental chamber with the exhaust valve closed, and then exhaled through the outlet with the inlet valve closed. The dead space within the mask was minimal.

Lung function measurements

Parameters required for theoretical modelling of deposition include the breathing rate and lung capacity (forced vital capacity or FVC). These were measured at the QUT Health Services Centre during the week of the inhalation test. A spirometer was used to determine the FVC, and the breathing rate was found by observing the number of breaths in 30 s while pretending to count the subjects heart rate (to avoid the problem of the subject ‘controlling’ their breathing rate.) Breathing rate measurements were also attempted at the time of the test by observation of the subjects’ throat and chest movement, however some rates could not be determined due to the difficulty in detecting chest movement during relaxed nasal breathing. Thus the rate results from the Health Centre (measured for normal breathing) were used as they were found to be similar to those few measured during the tests.

Experimental procedure

Aerosols were introduced into the large chambers to give a total number concentration of between 3 and 5x10^4 particles.cm^{-3}. The concentration was measured with the SMPS immediately prior to each test, and if necessary, was reduced to the required range by flushing the chamber with laboratory air. The sampling box was flushed with HEPA filtered air between different aerosol tests.

The mask was fitted to the subject and a simple leakage test was performed. The subject was requested to block off the exhalation tube and then to exhale slowly. If the exhaled air could be felt leaking out, they adjusted the tightness and position of the mask until this no longer occurred and the mask appeared to 'inflate' on their face, indicating no leakage.

To relax the subjects during the inhalation tests, they were asked to bring and read some material of their. Once the mask was fitted, the subject began reading. After breathing laboratory air through the mask for about five minutes to accustom them to the use of the mask, the subjects breathed nasally, inhaling aerosol from one of the chambers while exhaling into the sampling box for a period of approximately 10 minutes. During this time, the concentration in the chamber was measured (3 samples), followed by the exhaled aerosol from the sampling box (4 samples). The source concentration (chamber) was then measured again immediately after the test (3 samples).

The test was repeated for the second aerosol (from the second chamber), during which time the first chamber was flushed with laboratory air and filled with the third aerosol. The third aerosol was then tested giving a total testing time of not more than 1 hour for each subject.

Aerosol generation and characteristics

Methods

ETS

A smoke generator (described in (Morawska *et al.*, 1997) was used to generate the ETS. Only the side-stream smoke was used in this study by slowly pumping the smoke laden air from the smoke generator's chamber into the aerosol chamber. The resulting size distributions had count median diameters (CMD) of 0.183 μm with GSD of 1.7. An example of a typical ETS size distribution measured in the aerosol chamber is presented in Figure 2a. A concentration of approximately $5x10^4$ particles.cm^{-3} was achieved in the 3 m^3 chamber by smoking one cigarette.

Diesel smoke

The engine of a diesel vehicle parked outside the laboratory building (Toyota Land Cruiser) was allowed to idle for a few minutes. A large plastic bag of volume approximately 70 L was used to take a sample approximately 1 m from the exhaust pipe. The bag was then taken to the laboratory and emptied into the aerosol chamber. The time between taking the sample and delivering it into the chamber was approximately five minutes. The resulting size distributions had CMD of 0.125 μm with GSD of 1.7 An example of a typical diesel exhaust size distribution measured in the aerosol chamber is presented in Figure 2b. A concentration of approximately $5x10^4$ particles.cm^{-3} was achieved in the 1 m^3 chamber from one full bag.

Petrol smoke

A petrol generator was started and allowed to idle for a few minutes. A sample was taken from the exhaust using a large plastic bag of volume approximately 70 L. The bag was emptied into the aerosol chamber. The time between taking the sample and delivering it into the chamber was approximately one minute. The resulting size distributions had CMD of 0.069 μm with GSD of 1.7. An example of a typical petrol exhaust size distribution measured in the aerosol chamber is presented in Figure 2c. A concentration of approximately 5×10^4 particles.cm^{-3} was achieved in the 1 m^3 chamber from a half full bag.

It should also be noted that the CMD's of the aerosols generated for the purpose of this study, was in general larger than of the same type of aerosols measured in field studies or directly from the exhaust. For example ETS measured in a club had a CMD of approximately 0.067 μm (Morawska *et al.*, 1997); diesel measured directly from an exhaust at idle speed was approximately 0.063 μm (Morawska *et al.*, 1998), and petrol ranged from 0.039 – 0.060 μm (Ristovski *et al.*, 1998). The explanation to these differences is different for ETS and for the vehicle exhaust aerosol. Size of the ETS originating from the generator has been shown to be larger than this originating from a human smoker (Morawska *et al.*, 1997). Larger sizes of diesel and petrol aerosol can be explained as a result of rapid coagulation taking place in the bag immediately after collection from vehicle exhaust and prior to delivering it into the aerosol chamber. Initial concentrations in the bag (of the order of 10^6 particles.cm^{-3}) are high enough to support this hypothesis. The differences between field and laboratory generated aerosol was not a concern in this study. This is because firstly, it has been shown in the literature that there is a large variation in size distributions for aerosol generated from the same type of source, but measured in different studies;and secondly, the size distribution was actually measured in this study, and therefore no assumptions needed to be taken about its characteristics.

Reproducibility

There was some degree of variability in each aerosol spectrum from test to test. For ETS, the CMD's had a standard deviation (SD) of 0.030 μm, for diesel the SD was 0.025 μm and for petrol 0.017 μm. This variability was most likely caused by factors such as change in outside temperatures (when collecting diesel samples, for example, the engine could initially be colder), or variation in humidity (allowing more or less coagulation of particles in the collection bag). Also the time taken to deliver the diesel and petrol aerosols into the sampling chamber varied by the order of a minute or two, thus allowing more or less coagulation of particles. The above variations in CMD's however, would not affect the depositions observed in this study because each aerosol had little variation with respect to both concentration and size distribution throughout each individual subject's test.

Losses due to deposition in system

Losses of particles due to deposition in the tubing, mask and sample chamber were reported by Morawska et al, 1999 to be very low. To minimize the losses even further the spiral-walled type tubing used by (Morawska *et al.*, 1999) was replaced by shorter lengths of smooth-walled plastic tubing. The deposition was estimated to be approximately 2%, down from 7% in the previous study. The losses of particles in the system were thus considered to be small compared with other factors, in particular, intra-subject variability (measured to be ±11.5% by (Morawska *et al.*, 1999)).

Data analysis methods

Total particle number concentrations

The percent of aerosol deposited in each subjects' lungs was calculated by examining the total number concentrations of the source and exhalation sample for each aerosol.

The total number concentrations for each of the three initial and three final source samples, and for each of the four exhalation samples were extracted from the SMPS data. A graph of these total number concentrations was plotted for each aerosol inhaled by each subject. Initially, to accurately reflect the decrease in source concentration over the time of measurement, an exponential decay curve was fitted to the source data. This allowed the interpolation of source concentrations at the corresponding times of the exhalation measurements. It was found however, that a straight line fit was similarly representative of the decay in source concentration over the short measurement period and this simpler method of calculation was thus used to interpolate source concentration values. The percent deposition at each exhalation measurement was calculated, and then the average and standard deviations were found.

Fractional deposition

Data from the SMPS, which consists of the number concentration of particles in each of 102 SMPS size fraction channels was used to determine the fractional deposition for each subject and aerosol. Since the method used for calculation of the total number deposition was rigorous but labour intensive, a simpler method was tested on the total number concentration data using averages rather than linear interpolation. This method was found to give resulting depositions within 2% of the rigorously calculated values. As there are substantially more data involved in fractional deposition calculations, the simpler method was adopted for the calculations. The summary of this method is as follows. For each size fraction, the source concentration was estimated to be the average of all the source concentrations measured both before and after the inhalation test. The four exhaled concentrations were averaged, to provide a single exhalation concentration value. The difference between the source and exhalation values was then divided by the source concentration and converted into a percentage, to give the deposition in that particular size fraction. This was repeated for each of the 102 size fractions, for each subject and for each aerosol.

The number of particles in the extreme lower and upper size fractions of each aerosol was relatively small when compared with the peak of the distribution (either background noise or the tail of the normal distribution). As data from these extremes gave unrealistic and widely varying fractional depositions, the range was reduced to within two standard deviations of the mean of the distribution (representing 95% of the data). This was done by fitting a Gaussian distribution curve to 14 random samples of each aerosol to determine the average lower and upper size limits. These limits were then applied to the resulting fractional deposition graphs for each aerosol.

Results and discussion

Total number deposition

The percentage depositions for each subject and each aerosol are presented in Table 1. The average total number deposition of ETS was 36% with the SD of 10%. For diesel smoke, the average was 30% with SD of 9%, and petrol smoke, 41% and 8% respectively.

The inhaled and exhaled CMDs of each aerosol are very similar, indicating that there is no significant change in the particle size distribution after aerosol residence in the lung. It cannot be concluded however, that there was no growth due to condensation occurring in the lungs. Certain extent of particle growth by condensation in the humid environment of the lung is expected, and for example for ETS, a computer model by (Schroeter *et al.*, 2001) predicts an 80% increase in the particle diameters for all particles in this size range, on exiting from the nasopharyngeal region. In this study the aerosol undergoes several changes in temperature and humidity throughout the inhalation and measurement process; from relatively low humidity and temperature in the chamber, to high temperature and humidity in the lung, and returning to low humidity and temperature in the sampling box and then in the SMPS. Therefore a simple comparison of the measured inhaled and exhaled CMDs does not allow for conclusions as to the changes to particle characteristics whilst in the lung.

The respiratory rates were slightly higher than would be expected from a relaxed breathing state, probably due to being measured under conditions closer to a ‘normal’ (at the Health Services clinic) rather than a ‘relaxed’ breathing pattern.

The relationships between CMD, aerosol type and total number deposition are shown in Figure 3. Inspection of the results presented in Figure 3 gives an indication of, not only the variability of the total number deposition with aerosol type, but also of the inter–subject variability. For example, within the petrol smoke CMD range of approximately 0.060 to 0.080 µm, the total number deposition ranges randomly from about 30 to 50%, indicating an inter–subject variability of around 20%.

Total number deposition versus physiological factors

The average deposition of all three aerosols in each subject (person) was calculated and graphed against Age, Lung Capacity, and Respiratory Rate in Figures 4 a, b and c, respectively. The equation derived from the fitted regression is shown in each case. It can be clearly seen that increasing lung capacity is significantly predictive of increased deposition (P=0.03), with each unit increase in deposition estimated by an increase of the magnitude of 4.35 for each unit of lung capacity. It can also be seen that deposition cannot be predicted by either of the other physiological factors when considered individually.

Fractional deposition

The resulting fractional deposition curves for all subjects and each aerosol are shown in Figures 5, 6 and 7. From a visual inspection of the graphs it can be derived that the deposition of ETS is more dependent on subject rather than size fraction. Petrol and diesel smoke however show a distinct trend with size, independent of subject.

Figure 8 is a comparison of the average fractional deposition for all subjects (from the data in Figures 5, 6 and 7) for each of the three measured aerosols. The curves presented in Figure 8 for each aerosol show the differences in deposition due to characteristics other than size, for each aerosol. Error bars have not been included in Figure 8 for reasons of clarity, however the approximate standard deviations were 12% for ETS (range 39%), 11% for diesel smoke (range 38%), and 10% for petrol smoke (range 35%).

Figure 8 also shows that the average deposition of ultrafine particles (particles <0.1 μm) is greater than 30% for both diesel and petrol smoke. Deposition of particles in this range show exceptional toxicity with regards to lung morbidity and mortality (Donaldson et al, 1998).

For comparison, Figure 9 shows data from the present study, super-imposed upon curves produced from (Stahlhofen *et al.*, 1989); (modelling of experimental data from two other authors). Although Figure 9 shows mouth breathing deposition, a nasal deposition curve from (Heyder *et al.*, 1986) (whose data from three subjects was used in the model by (Stahlhofen *et al.*, 1989) is also comparative to the given curves. Thus the above curves may be used to give some comparison of our results with previous studies. It was expected that the data from the present study would lie somewhere within the range (on the y-axis), of that given in Figure 9, as the breathing frequencies and flow rates measured here were those of a relaxed person at rest. Whilst this is true for petrol and diesel smoke, it is not the case for ETS. The x-axis of Figure 9 represents the diameter of unit density spheres, however the actual aerosols measured for some of the data were monodisperse di-2-sthylhexyl sebacate, iron oxide and silver aerosols (Heyder *et al.*, 1986). Given the different aerosol type here (polydisperse ETS, petrol and diesel smoke) a shift in diameter would be expected for this data. This is seen for diesel smoke where there is a minimum in the curve at 0.23 μm, whereas the minimums for the given curves are around 0.37 μm. It could be assumed that the ETS and petrol data are also shifted by an amount reflecting the density and shape of the aerosol particles.

Statistical Analysis

Overall Fits

The data from figures 5, 6 and 7was examined to give an indication of inter-subject variability and aerosol deposition pattern variability. The results of overall linear fits are shown in Table 2, where t is the test statistic corresponding to the null hypothesis that the coefficient is equal to zero (ie, if the slope is zero then there is no relationship between that aerosol and the response). The p-value is the corresponding probability of obtaining the estimated coefficient or a more extreme estimate if the null hypothesis is true. R-squared is the proportion of total variation in the response that is explained by the regression model.

For petrol smoke, both the intercept and the slope are highly significant (p-values less than 0.0000). The R – squared value indicates a much better linear fit to the data for petrol than for diesel. As expected from visual inspection of the data in Figure 6, diesel is better explained by a quadratic relationship:

$$Diesel = 47.80 - 209.48\,X + 465.41\,X\hat{}2$$
$$R\text{-}square = 0.1637$$

Whereas the intercept of the linear fit to the ETS data is significantly different from zero, the corresponding slope is not significantly different from zero (p=0.3492). This indicates that ETS fractional deposition is independent of particle diameter. No other substantially better relationship could be found for ETS.

Test of Equality of Slopes

A test of the hypothesis of equal slopes of the linear relationships for petrol, diesel and ETS revealed (after accounting for individual variation), that there is a significant difference in the straight-line patterns of the three exposures over particle size (the ANOVA test statistic F = 4.31, p = 0.016).

Subsequent pairwise analyses revealed significant differences between the slopes for petrol and diesel (p=0.0003) and petrol and ETS (p=0.0000) but no significant difference between the slopes for diesel and ETS (p=0.35). However, this latter result should be considered in light of the observation above that diesel is better described by a quadratic rather than a linear pattern.

Test of Individual Patterns

Comparisons were made between individuals' distributions of petrol, diesel and ETS over particle size. For each of the 14 individuals, straight-line regressions were fit to each exposure and a test of equality of the three slopes was then performed. The following results were obtained:

- For petrol, all 14 individuals showed a significantly linear, negative pattern over particle size (thirteen p-values less than 0.1%, one p-value less than 1%).
- For diesel, 9 out of the 14 individuals showed a significantly linear, negative pattern. Two individuals had positive slopes but these were not significant. (eight p-values less than 0.1%, two p-values less than 1%, one p-value less than 5%). Three individuals showed a linear slope but it was not significantly different from zero.
- For ETS, 9 out the 14 individuals demonstrated no significant linear pattern (5 negative slopes, 4 positive slopes); 4 individuals demonstrated a significantly linear, negative pattern (two p-values less than 0.1%, two p-values less than 1%) and one individual showed a significant, positive pattern (p-value less than 1%).
- When comparing the deposition patterns of the three aerosols for each individual, 12 out of the 14 showed there was a significant difference between each aerosol, whilst there was no significant difference for the remaining 2 individuals.

Conclusions

In this study the number concentration and size distribution of combustion aerosol particles in the submicrometer range inhaled and exhaled by 14 human volunteers was experimentally measured to establish total submicrometer and size fractional deposition in the particles in the respiratory tract. The investigations included ETS aerosol, diesel and petrol emissions. The average total number deposition of ETS was 36% with a SD of 10%. For diesel smoke, the average was 30% with SD of 9%, and petrol smoke, 41% and 8% respectively. The study showed that increasing lung capacity is significantly predictive of increased deposition (P=0.03), however, the deposition cannot be predicted by either of the other physiological factors when considered individually (age in the range from 20 to 30 years or respiratory rate). In the analysis of size dependent deposition, the following conclusions were derived from the study:

- Petrol smoke deposition pattern is best represented by a linear fit with a significant intercept and a significant negative slope, ie there is a constant change in deposition with diameter.
- Diesel smoke deposition pattern is best represented by a quadratic equation, with a minimum deposition occurring at around 0.23 μm.
- ETS deposition pattern is best represented by a linear fit with a significant intercept and a non-significant slope, indicating no dependence of deposition on diameter.
- Intra-subject and inter-subject variability were compared in the assessment of equal intercepts and slopes. Thus, even after accounting for individual variation, there is a significant difference in the straight-line patterns of the three types of aerosol over particle size.
- The analysis of the deposition patterns of all three aerosols in each individual shows that there is a significant difference between each aerosol for a majority of individuals (12 out of the 14). This is an important result as it indicates that differences persist regardless of inter-subject variability.

Results from (Morawska *et al.*, 1999) showed a total number deposition for nasal breathing of ETS to be 56 ± 15.9%, which is higher than 36% (SD 10%) found in this study. Both studies, however, showed a similar trend in the fractional deposition curves which were linear with a zero slope. Whilst a similar experimental set up was used in both cases, some small changes need to be noted. For this study:

- smooth walled plastic tubing was used to replace the spiral tubing of slightly higher deposition rating;
- a different brand of cigarette was used;
- different subjects were used from the previous study;
- three samples of source aerosol both before and after the four exhalation samples were measured as opposed to one source sample before and after five exhalation samples previously.

Although total ETS deposition measured in this study was smaller than that found in the earlier experiments (36% vs. 56%), the measured total deposition values for all three test aerosols are still consistently higher than the corresponding theoretical predictions, with ETS particles exhibiting the greatest difference. Calculated average total deposition values for all 14 volunteers, considering the specific anatomical and respiratory parameters of each volunteer and the specific size distribution for each inhalation experiment, were 16.5% for ETS, 20.2% for diesel, and 29.9% for petrol particles (compared to 36.2%, 29.6%, and 41.1% in the present experiments). To bridge the gap between experimental and theoretical data, additional physical mechanisms, acting primarily on non-spherical chain aggregates, such as interception, or mechanisms reducing particle size upon inspiration, such as evaporation of semi-volatile compounds, were implemented into the stochastic deposition model. The effect of these mechanisms on total deposition and the comparison of the revised predictions with the experimental data presented in this study is presented elsewhere (Hofmann *et al.*, Submitted for publication).

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			Resp.	*diesel*				*ETS*				*petrol*			
		FVC	Rate	*Deposition*		*CMD (μm)*		*Deposition*		*CMD (μm)*		*Deposition*		*CMD (μm)*	
Subject	**Age**	**(L)**	**(/min)**	*%*	*SD*	**Inhaled**	**Exhaled**	*%*	*SD*	**Inhaled**	**Exhaled**	*%*	*SD*	**Inhaled**	**Exhaled**
201	28	4.04	16	25	0.7	0.138	0.142	37	1.8	0.190	0.189	36	2.2	0.076	0.079
202	21			36	1.0	0.127	0.132	44	1.3	0.205	0.209	51	2.3	0.064	0.073
203	27	5.42	18	40	2.9	0.187	0.189	52	1.8	0.196	0.202	50	3.7	0.117	0.131
204	21	5.80	16	43	6.8	0.146	0.144	44	1.9	0.210	0.212	51	1.8	0.072	0.078
205	24	3.83	12	25	1.8	0.139	0.138	27	1.1	0.177	0.175	36	2.8	0.063	0.063
206	27	4.63	22	32	1.3	0.101	0.104	35	1.9	0.140	0.142	43	0.7	0.057	0.057
207	28	3.76	16	25	2.6	0.100	0.103	21	1.6	0.153	0.154	36	1.5	0.070	0.071
208	29	5.70		33	3.7	0.111	0.110	38	4.8	0.174	0.169	42	4.5	0.073	0.076
209	20	3.29	17	17	1.4	0.122	0.128	53	1.3	0.177	0.177	34	3.3	0.062	0.069
210	28	4.01	20	38	4.4	0.097	0.105	21	2.7	0.156	0.159	29	2.3	0.087	0.094
211	20	3.88	16	35	2.2	0.124	0.128	29	1.0	0.217	0.218	42	0.7	0.063	0.069
212	20	5.14	18	20	1.1	0.128	0.127	29	0.5	0.224	0.222	36	1.3	0.059	0.062
213	20	5.32	18	13	1.4	0.126	0.127	36	0.6	0.212	0.208	33	1.6	0.060	0.064
214	26	6.37	20	35	2.1	0.087	0.092	41	1.1	0.130	0.131	57	1.7	0.043	0.048
Averages:	24	4.71	17	30		0.124	0.126	36		0.183	0.183	41		0.069	0.074
SD:				9		0.025	0.024	10		0.030	0.030	8		0.017	0.020

Table 1. **Deposition of particles using the total number concentration for each of the 14 subjects and three aerosols.**

Aerosol	Coefficient	s.e.	t = (coef/se)	p-value	R-squared
Petrol					0.45
Intercept	53.65	0.681	78.8	0.0000	
Slope	-158.7	6.174	-25.7	0.0000	
Diesel					0.052
Intercept	35.15	0.073	45.5	0.0000	
Slope	-28.36	4.153	-6.83	0.0000	
ETS					0.001
Intercept	36.72	0.750	48.97	0.0000	
Slope	-2.36	2.523	-0.937	0.3492	

Table 2. Overall linear fits. Where s.e. is the standard error, t is the test statistic, p-value is the corresponding significance level, and R-squared is the proportion of variation explained by the model.

Figure Captions

Figure 1. Equipment set-up showing subject breathing aerosol from $3m^3$ chamber.

Figure 2. Examples of size distribution spectra of (a) ETS, (b) diesel, and (c) petrol.

Figure 3. Relationship between CMD aerosol type and total number deposition.

Figure 4. Average total number deposition for each subject versus: (a) age in years, (b) Forced Vital Capacity (Lung Capacity), and (c) Respiratory Rate.

Figure 5. Fractional deposition of ETS for all subjects.

Figure 6. Fractional deposition of diesel smoke for all subjects.

Figure 7. Fractional deposition of petrol smoke for all subjects.

Figure 8. The average fractional deposition of all subjects for each of the three aerosols.

Figure 9. Total deposition for mouth breathing as a function of the diameter of unit density spheres, for a volumetric flow rate of 250 cm^3s^{-1} and breathing frequencies of 3.75/min and 15/min and tidal volumes of 2000 cm^3 and 500 cm^3 respectively. The curves represent the sum of the approximations of the mean regional depositions derived in the paper by Stahlhofen et al (1989). The average fractional deposition of all subjects for each of the three aerosols (from Figure 10 of this paper) have been included on this graph for comparison.

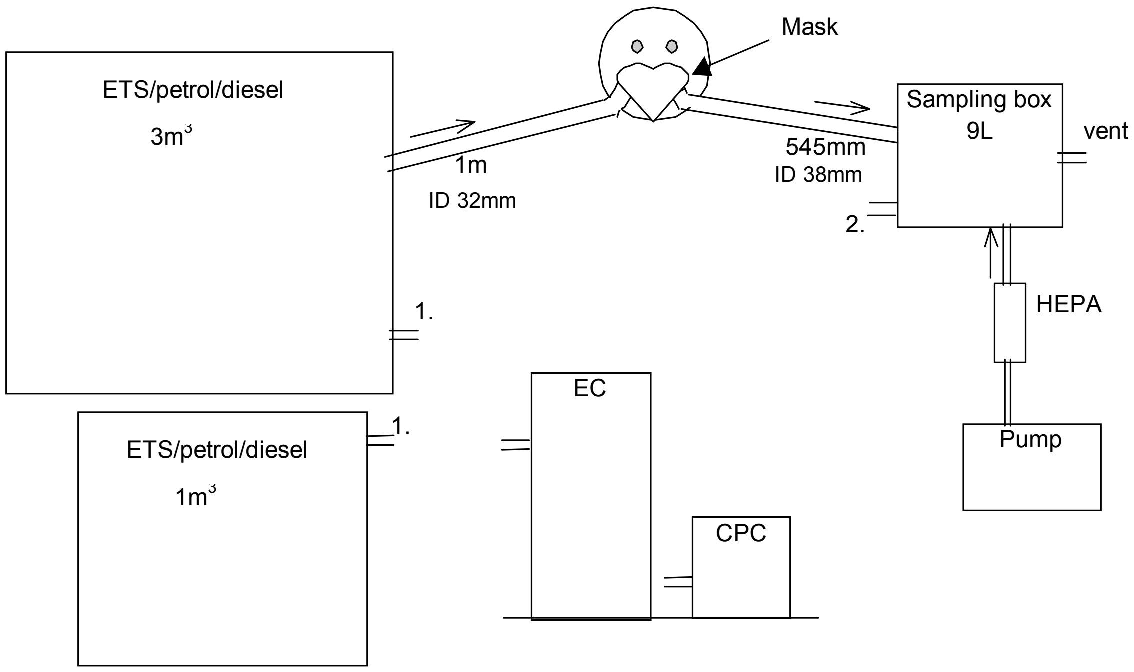


Figure 1. Equipment set-up showing subject breathing aerosol from $3m^3$ chamber.

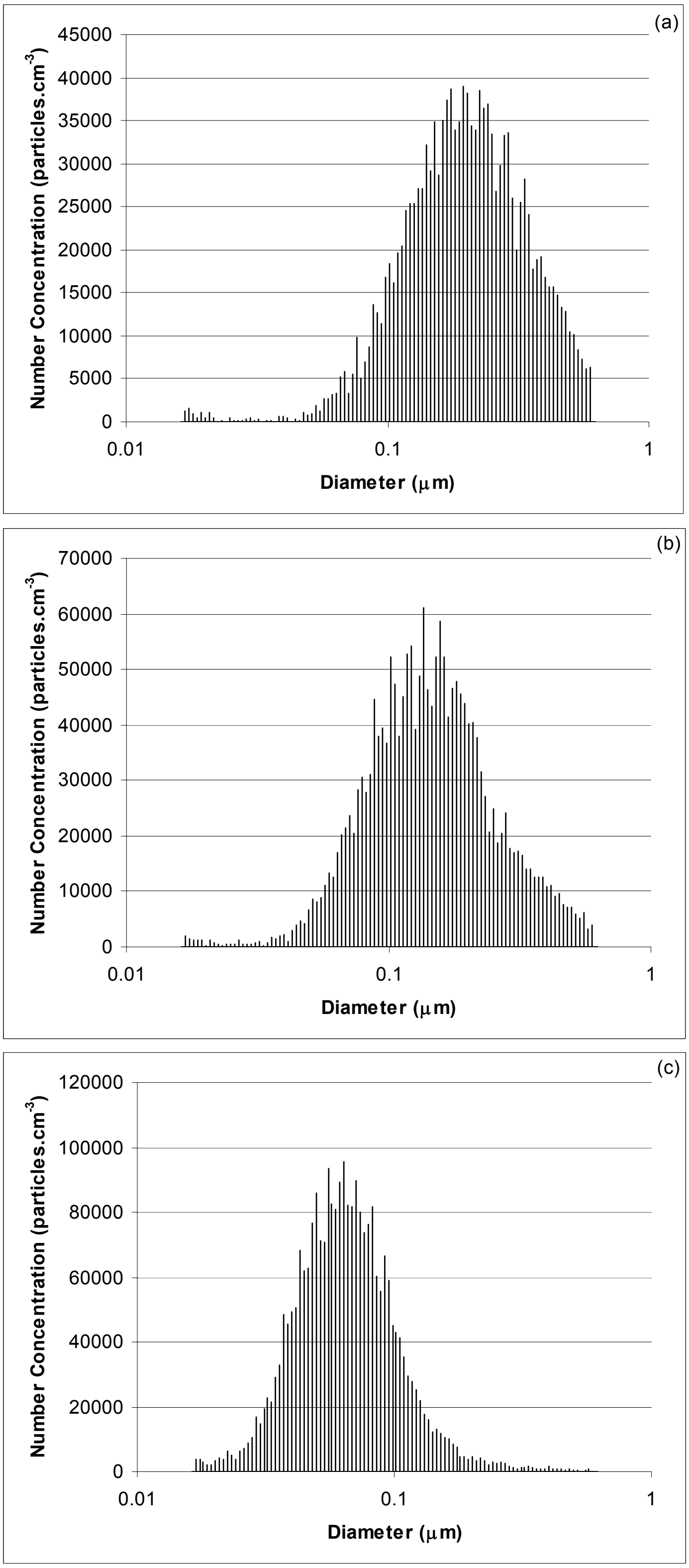


Figure 2. Examples of size distribution spectra of (a) ETS, (b) diesel, and (c) petrol.

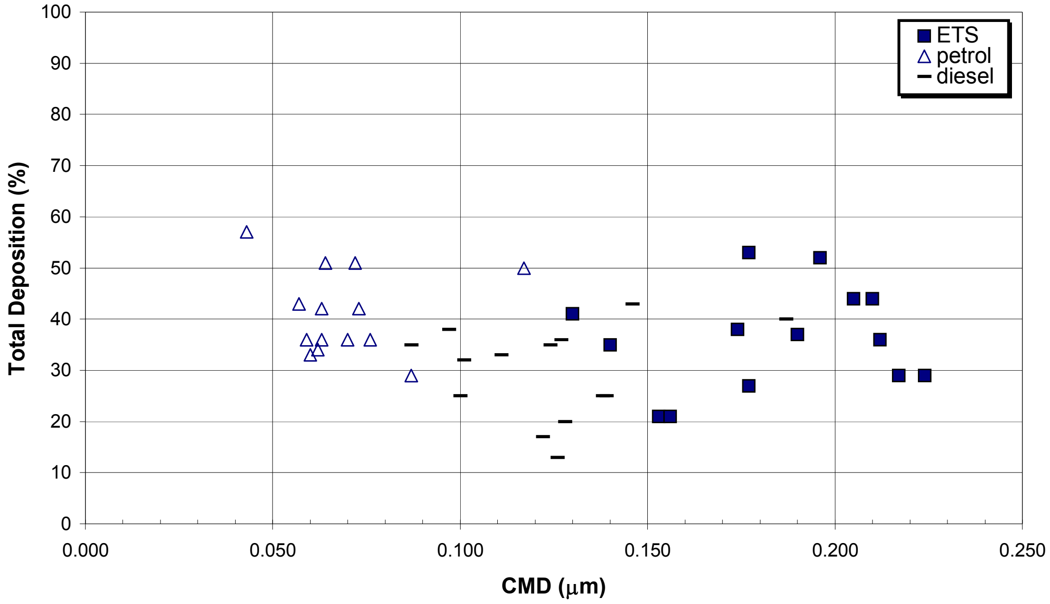


Figure 3. Relationship between CMD aerosol type and total number deposition.

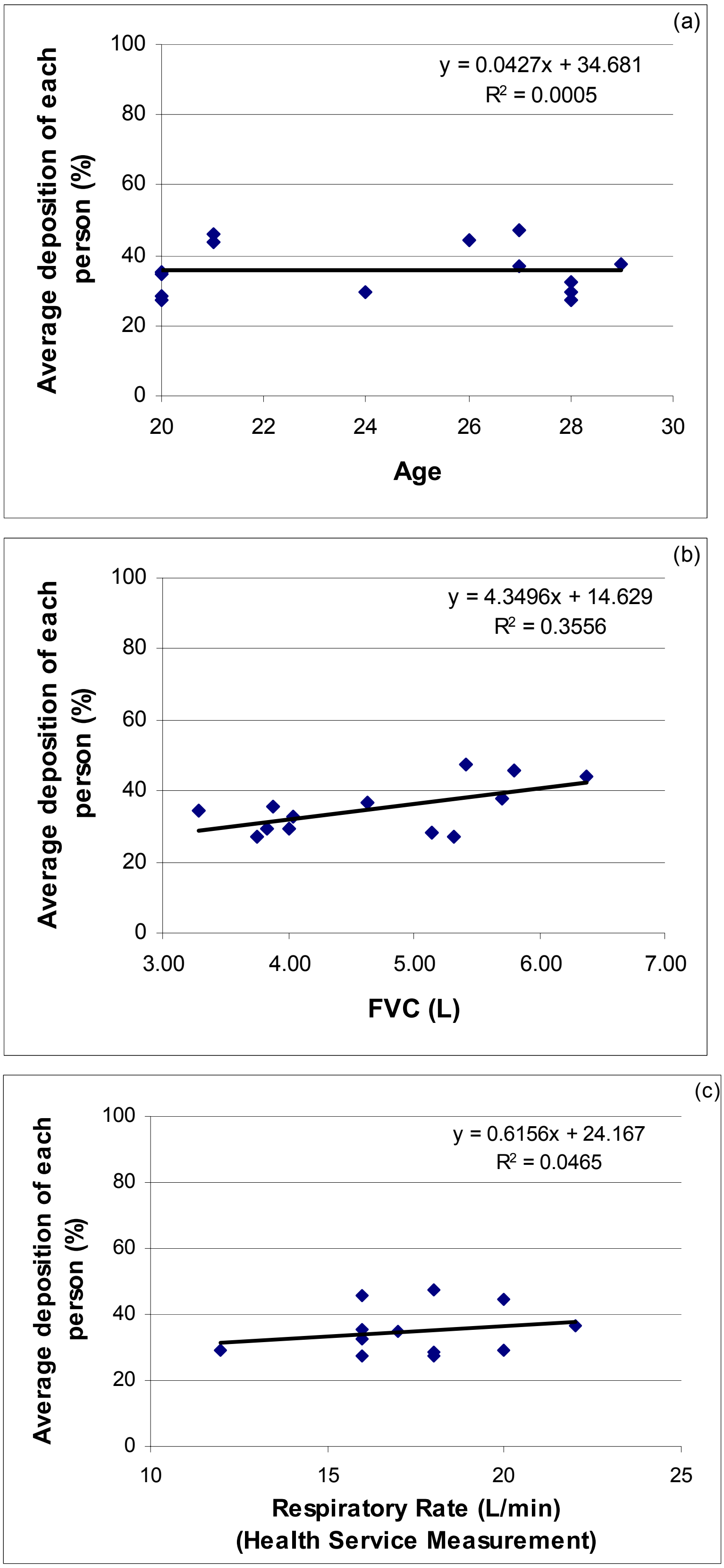


Figure 4. Average total number deposition for each subject versus: (a) age in years, (b) Forced Vital Capacity (Lung Capacity), and (c) Respiratory Rate.

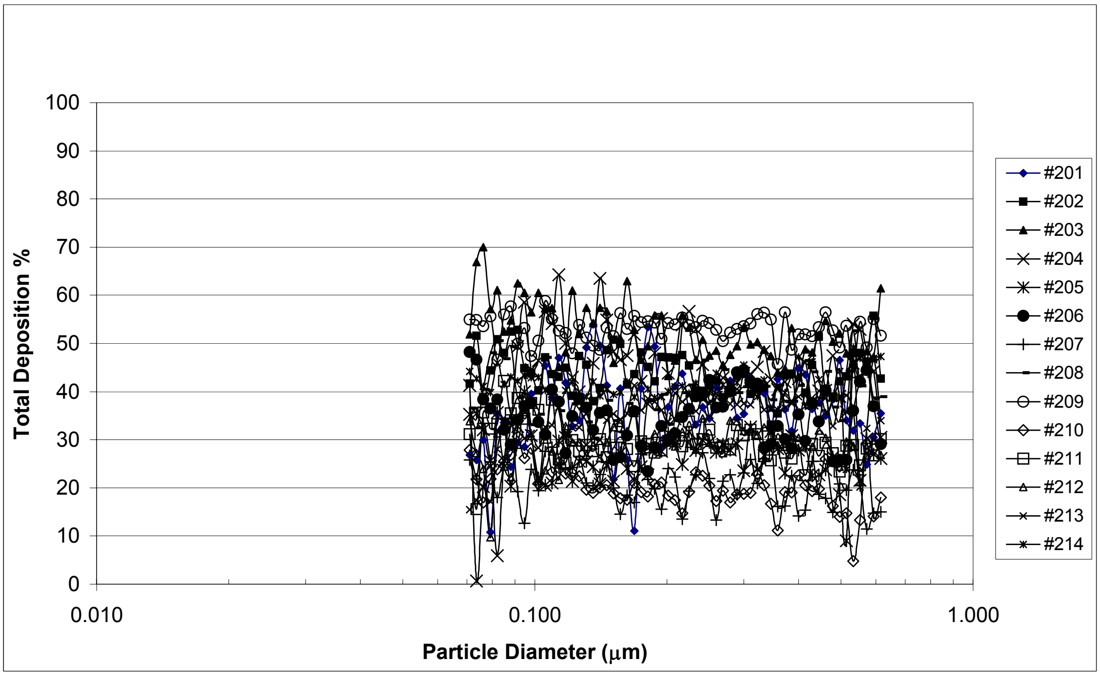


Figure 5. Fractional deposition of ETS for all subjects.

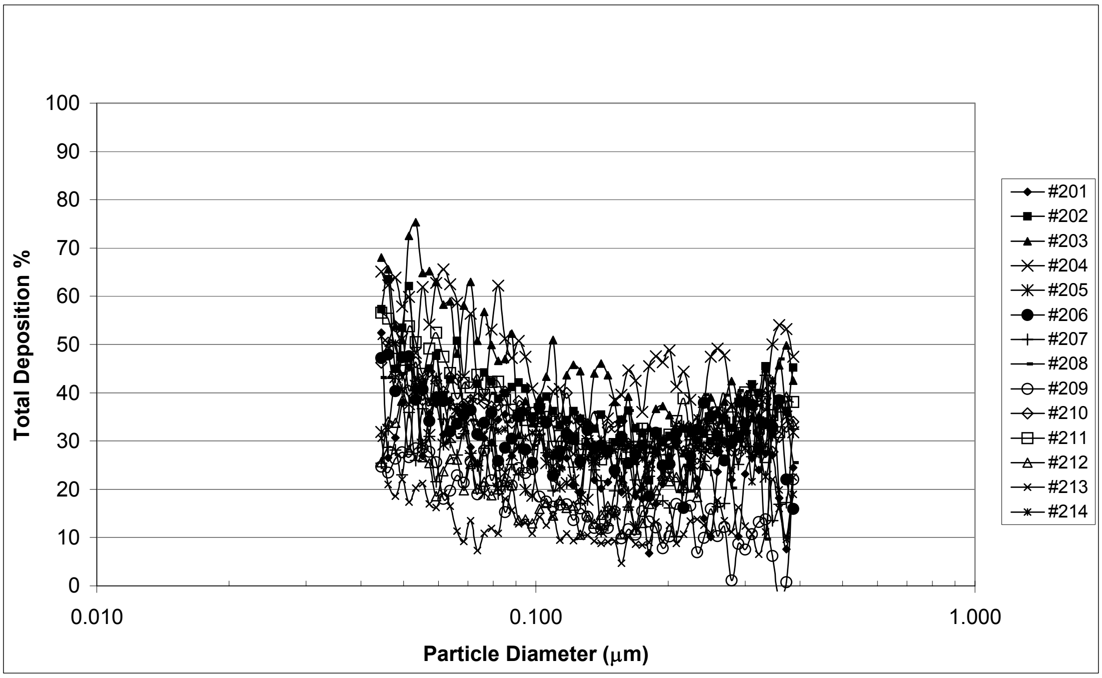


Figure 6. Fractional deposition of diesel smoke for all subjects.

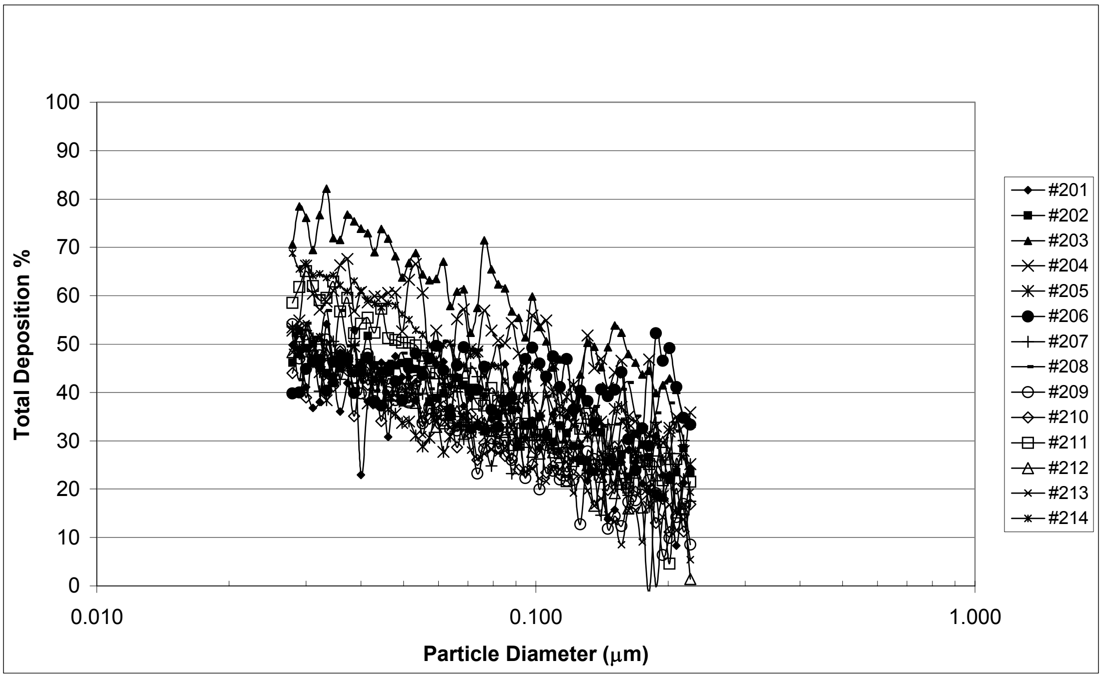


Figure 7. Fractional deposition of petrol smoke for all subjects.

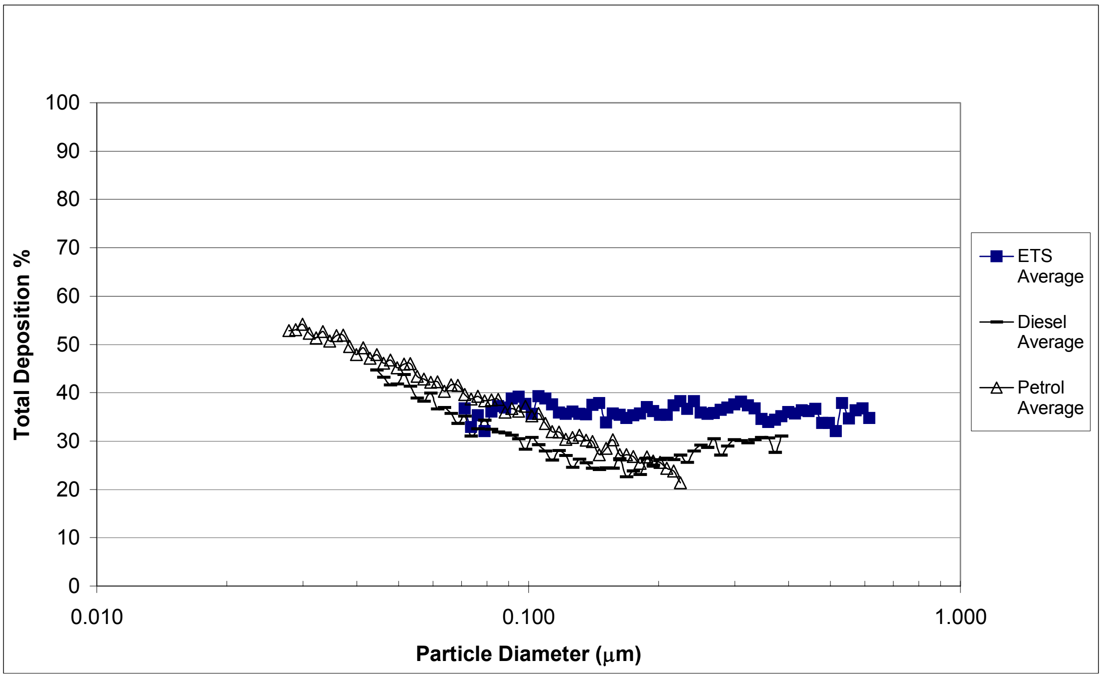
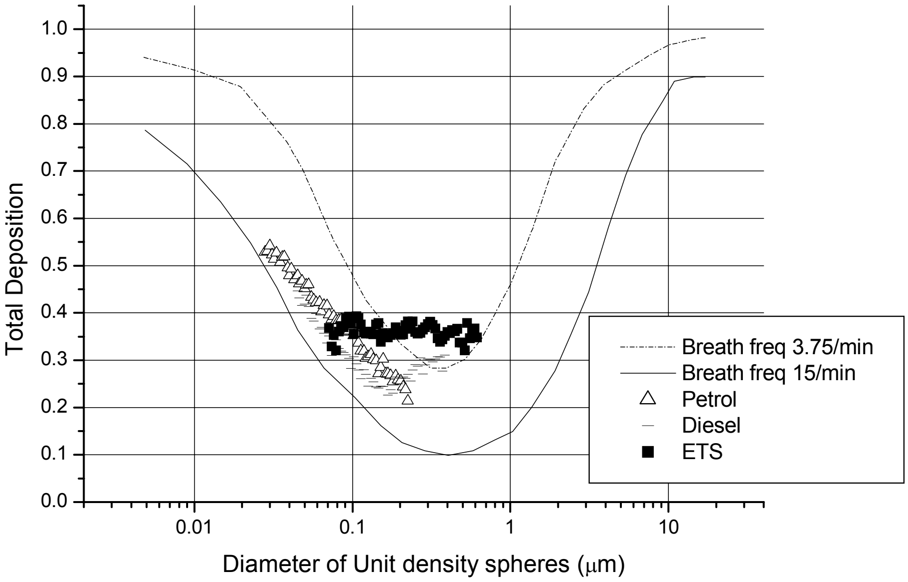


Figure 8. The average fractional deposition of all subjects for each of the three aerosols.



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